

**The 14th China-Japan Symposium on
Ferroelectric Materials and Their Applications
(CJFMA14)**

Kanazawa, Japan

December 8-9, 2022

Abstract & Technical Book

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CJFMA

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CJFMA14 Welcome Message

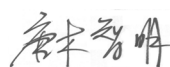
The 14th China-Japan Symposium on Ferroelectric Materials and Their Applications (CJFMA14) will be held in a hybrid format from December 8 to 9, 2022. The main venue is in Kanazawa, Japan. On behalf of the Organizing Committee, it is my great pleasure to welcome you to this exciting event.

The CJFMA1 was held from August 19 to 22, 2009 in Dunhuang, China. Since the first Symposium, our CJFMA was successfully held annually in Japan and China. The CJFMA2 was in Kurobe, Japan, the CJFMA3 in Xiangtan, China, the CJFMA4 in Matsushima, Japan, the CJFMA5 in Suzhou, China, the CJFMA6 in Yamanashi, Japan, the CJFMA7 in Beijing, China, the CJFMA8 in Tsukuba, Japan, the CJFMA9 in Chengdu, China, the CJFMA10 in Inuyama, Japan, the CJFMA11 in Nanjing, China, the CJFMA13 (hybrid format) in Shenyang, China. The 2021 International Workshop on New Domain Technology of Ferroelectric Materials and Their Applications (NDTFMA2021) was held online instead of the CJMAF12 by Toyama Prefectural University, Japan, because of the pandemic outbreak of Covid-19.

Ferroelectric materials, including piezoelectric materials, dielectric materials, microwave materials, electro-optical materials, electro-caloric materials, and other related materials, are an important class of functional materials that are widely used in our modern society. New materials, fabrication methods, evaluation methods, devices, applications, simulations, and theories are also the content of this Symposium.

Japan and China are the two most active countries in the world for ferroelectric materials research. The CJFMA is a platform for high-level academic exchange and deepening friendship between the two countries in the field. More young researchers and students are expected to participate our Symposium.

We are very pleased to have 73 contributions (2 plenary talks, 40 invited talks, and 31 posters) who submitted their current work to present at this hybrid conference. We think, this conference will surely be a memorable, successful and grand meeting.



Tomoaki Karaki (Zhiming Chen), Dr.
The Chairperson of CJFMA14

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CJFMA14 Organizing

Conference Chair:

Tomoaki Karaki (Zhiming Chen) (Toyama Prefecture University, Japan),

Vice Chair:

Guorong Li (Shanghai Institute of Ceramics, CAS, China)

Advisory Committee:

Xi Yao (Xi'an Jiaotong University, China), T. Tsurumi (Tokyo Institute of Technology, Japan), Cewen Nan (Tsinghua University, China), S. Wada (Yamanashi University, Japan)

Academic Committee (arrange in alphabetical order of the surname):

Japan side: J. Akedo, A. Ando, R. Aoyagi, D. Fu, T. Fujii, T. Hoshina, K. Kakimoto, T. Karaki, Y. Kuroiwa, N. Fujimura, W. Sakamoto, H. Takeda, E. Tokumitsu, S. Tsukada, R. Wang, W. Wang, M. Yamaguchi, T. Yoshimura,

China side: Dinghua Bao, Xiangming Chen, Yanfeng Chen, Jinrong Cheng, Shuxiang Dong, Haoshuang Gu, Dong Guo, Yunbin He, Li Jin, Guorong Li, Jiangyu Li, Jingfeng Li, Hanxing Liu, Junming Liu, Shengguo Lu, Haosu Luo, Xiangjian Meng, Jun Ouyang, Wei Ren, Chunlei Wang, Hong Wang, Jinbin Wang, Yaojin Wang, Zhanjie Wang, Zhuo Xu, Tongqing Yang, Manwen Yao, Jiwei Zhai, Dou Zhang, Jialiang Zhang, Yichun Zhou, Jianguo Zhu, Kongjun Zhu

Poster presentation award judging committee

Chair: Eisuke Tokumitsu (JAIST)

Vice Chair: Haosu Luo (Shanghai Institute of Ceramics, CAS, China)

Members: Jianguo Zhu (Sichuan University)
Jun Ouyang (Qilu University of Technology)
Zhigao Hu (East China Normal University)
Tadashi Fujii (Toyama Prefectural University)
Wensheng Wang (Fujitsu Semi. Memory Solution Ltd.)

Local team:

Tomoaki Karaki (Zhiming Chen) (Toyama Prefectural University), Chair

Eisuke Tokumitsu (JAIST), Program & Venue

Tadashi Fujii (Toyama Prefectural University), Secretariat

Wensheng Wang (Fujitsu Semi. Memory Solution Ltd.), Accountant

CJFMA14 Oral presenters Guidelines Summary

1. [Please enter the meeting room 10 minutes before the session starts.](#)
Zoom meeting ID: 805 936 4730, password: jcfma14
2. Check your speaker, microphone and camera.
3. Rename your screen name into **【Presentation number】**
4. Reply to the chair if your name is called during attendance confirmation.
5. Turn off your microphone and camera after the attendance confirmation.
6. When your presentation turn comes, the chair will call your presentation number or name.
7. Turn on your microphone, camera and start sharing your screen (presentation files) to begin your presentation.
8. During the Q&A session, the chair will pick up the audience who has questions.
9. Please turn off your microphone, camera and stop sharing your screen after the Q&A session.

Time limitation for each presenter	
Plenary talks	40 minutes (35 min. for talk and 5 min. for Q&A)
Invited talks	20 minutes (15 min. for talk and 5 min. for Q&A)

First bell	Second bell	Third bell
2 mins before the talk ends	The end of the talk	The end of the presentation

***Please make sure to finish your talk within the determined time period. Talk or Q&A might be stopped by the chair if the presenter overran the allotted time.**

*Please check out the Presenters Guidelines if you need more information:

https://www.pu-toyama.ac.jp/JCFMA14/Presenters_Guidelines.pdf

CJFMA14 Poster presenters Guidelines Summary

1. Please enter the meeting room 15 minutes before the session starts.
Zoom Meeting ID: 521 666 2126, password: jcfma14
2. Check your speaker, microphone and camera.
3. Rename your screen name into **【Poster number & last name】** .
4. Enter the breakout room named by your poster number & name.
5. Make the screen-shear to show your ppt.
6. When someone(s) come(s) into your breakout room, you can begin your presentation in English.
7. If someone(s) aske(s) questions in English or Japanese or Chinese, you can also answer in English or Japanese or Chinese.
8. The poster session period is 11:10-12:40 of Dec. 9, 2022 (Japan/Korea Standard Time).
9. You can leave the breakout room after 12:40, if finished all answers to questions.

*Please check out the Presenters Guidelines if you need more information:

https://www.pu-toyama.ac.jp/JCFMA14/Presenters_Guidelines.pdf

CJFMA14 Arrangement

<u>Dec. 8, 2022 (Japan/Korea Standard Time)</u>		
Opening Address	9:00-9:05	Tomoaki Karaki (Zhiming Chen)
Plenary Talks	9:05-9:45 (I-1)	Norihiko Oshime
	9:45-10:25 (I-2)	Jiangyu Li
Introduce of CJFMA15	10:25-10:35	Jun Ouyang
Break	10:35-10:45	
Invited Session A	10:45-11:05 (A-1)	Takeshi Yoshimura
	11:05-11:25 (A-2)	Hang Luo
	11:25-11:45 (A-3)	Yaojin Wang
	11:45-12:05 (A-4)	Jun Ouyang
	12:05-12:25 (A-5)	Yiping Guo
Lunch	12:25-13:30	
Invited Session B	13:30-13:50 (B-1)	Hiroaki Takeda
	13:50-14:10 (B-2)	Yuui Yokota
	14:10-14:30 (B-3)	Limei Zheng
	14:30-14:50 (B-4)	Muneyasu Suzuki
	14:50-15:10 (B-5)	Yongke Yan
	15:10-15:30 (B-6)	Yohachi Yamashita
	15:30-15:50 (B-7)	Chengtao Luo
Break	15:50-16:00	
Invited Session C	16:00-16:20 (C-1)	Nan Zhang
	16:20-16:40 (C-2)	Zhiguo Yi
	16:40-17:00 (C-3)	Jing Wang
	17:00-17:20 (C-4)	Zhengqian Fu
	17:20-17:40 (C-5)	Xiao Yu
	17:40-18:00 (C-6)	Yanxue Tang
	18:00-18:20 (C-7)	Jie Jiao
Banquet	19:00-20:30	

<u>Dec. 9, 2022 (Japan/Korea Standard Time)</u>		
Invited Session D	9:00-9:20 (D-1)	Bo-Ping Zhang
	9:20-9:40 (D-2)	Dawei Wang
	9:40-10:00 (D-3)	Hajime Nagata
	10:00-10:20 (D-4)	Zhang Jialiang
	10:20-10:40 (D-5)	Wataru Sakamoto
	10:40-11:00 (D-6)	Zhiyong Zhou
Break	11:00-11:10	
<u>Poster Session</u>	11:10-12:40	<u>P-01 – P-31</u>
Lunch	12:40-13:30	
Invited Session E	13:30-13:50 (E-1)	Yuji Noguch
	13:50-14:10 (E-2)	Hua Hao
	14:10-14:30 (E-3)	Zhigao Hu
	14:30-14:50 (E-4)	Satoshi Wada
	14:50-15:10 (E-5)	Masaki Yamaguchi
	15:10-15:30 (E-6)	Wei Ren
	15:30-15:50 (E-7)	Shinya Tsukada
Break	15:50-16:00	
Invited Session F	16:00-16:20 (F-1)	Laiming Jiang
	16:20-16:40 (F-2)	Jun Akedo
	16:40-17:00 (F-3)	Eisuke Tokumitsu
	17:00-17:20 (F-4)	Rao Badari Narayana
	17:20-17:40 (F-5)	Jundong Song
	17:40-18:00 (F-6)	Jing Yan
	18:00-18:20 (F-7)	Xueyou Yuan
	18:20-18:40 (F-8)	Lisha Liu
Closing Remarks	18:40-18:50	Tomoaki Karaki (Zhiming Chen)

List of Poster (11:10-12:40, Dec. 9, 2022)

No.	Name	Affiliation
P-01	Yao Su	Baoji University of Arts and Sciences
P-02	Yu Xie	Nanjing University of Aeronautics and Astronautics
P-03	Jinfeng Lin	Tongji University
P-04	Takayasu Shigemasu	Hiroshima University
P-05	Xiaonan Ma	Shanghai University
P-06	Mingyang Shao	Hiroshima University
P-07	Adil Emin	Lanzhou University
P-08	Xianfa Ai	Nanjing University of Aeronautics and Astronautics
P-09	Takumi Nozaki	Nagoya Institute of Technology
P-10	Zhe Wang	Xi'an Jiaotong University
P-11	Peimei Yuan	Baoji University of Arts and Sciences
P-12	Atsuto Fujisawa	Shizuoka University
P-13	Yu Xiang	Shonan Institute of Technology
P-14	Kohei Miyaji	Osaka Metropolitan University
P-15	Shuting Yang	Shandong University
P-16	Kazuto Yoshida	Shizuoka University
P-17	Soma Sakoda	Osaka Metropolitan University
P-18	Xiaojie Li	Shanghai University
P-19	Seiji Sogen	Shizuoka University
P-20	Weiwei Yang	Shanghai Institute of Ceramics
P-21	Hongrui Jia	Xi'an Jiaotong University
P-22	Jun Hirade	Shibaura Institute of Technology
P-23	Kun Zhu	Tongji University
P-24	Jin Qian	Tongji University
P-25	Ying Shi	Shanghai Institute of Ceramics
P-26	Fei Jing	Baoji University of Arts and Sciences
P-27	Tomohiro Nishimoto	Shizuoka University
P-28	Ruixuan Xue	Baoji University of Arts and Sciences
P-29	Bo Zhao	Baoji University of Arts and Sciences
P-30	Nina Dai	Baoji University of Arts and Sciences
P-31	Shicheng Peng	Tokyo Institute of Technology

PROGRAM

(Hybrid format)

Dec. 8, 2022 (Japan/Korea Standard Time)

9:00 Opening address, Tomoaki Karaki (Zhiming Chen)

Oral Session

9:05-10:35 Plenary Session Chair: Hiroaki Takeda (Saitama University)

9:05 I-1

Bragg Coherent X-ray diffraction for visualization of the inhomogeneous structure of a single ferroelectric particle

N. Oshime, K. Ohwada, K. Sugawara, A. Shimada, N. Fukushima, T. Ueno, A. Machida, T. Watanuki, S. Ueno, I. Fujii, S. Wada, K. Momma, K. Ishii, H. Toyokawa, S. Kim, S. Tsukada, and Y. Kuroiwa

National Institutes for Quantum Science and Technology, Japan

9:45 I-2

Polar orders across scales: the effect of strain and mechanics

Jiangyu Li

Southern University of Science and Technology, China

10:45-12:25 Invited Session A Chair: Yuui Yokota (Tohoku University)

10:45 A-1

Energy Harvesting from Electric Power Lines Using Piezoelectric Resonator

Takeshi Yoshimura, Shuichi Murakami, and Norifumi Fujimura

Osaka Metropolitan University, Japan

11:05 A-2

Improved capacitive energy storage nanocomposites at high temperature utilizing ultralow loading of bimetallic MOF

Hang Luo, Fan Wang, Xiaona Li, Dou Zhang

Central South University, China.

11:25 A-3

Ferroelectrics for emerging oral bioelectronics

Yaojin Wang, Xuehui Zhang and Xuliang Deng

Nanjing University of Science and Technology, China

11:45 A-4

High performance dielectric energy storage via nanograin engineering in ferroelectric/antiferroelectric film capacitors

Jun Ouyang, Hongbo Cheng, Xiao Zhai, Jing Yan, Yu Su

Qilu University of Technology, China.

- 12:05 A-5
Reducing the coercive field of rhombohedral NBT-based crystals by domain engineering
Geng Huangfu, Haosu Luo, Yiping Guo
Shanghai Jiaotong University, China.
- 13:30-15:50 Invited Session B, Chair: Wataru Sakamoto (Chubu University)
- 13:30 B-1
Growth and Characterization of $\text{Ba}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ Single Crystals
H. Takeda, T. Hoshina, and T. Tsurumi
Saitama University, Japan.
- 13:50 B-2
Effects of Al substitution on crystal structure and piezoelectric properties of $\text{Ca}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$ and $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ single crystals
Yuui Yokota, Yuji Ohashi, Takahiko Horiai, Akira Yoshikawa
Tohoku University, Japan.
- 14:10 B-3
Simultaneously achieving giant piezoelectricity and record coercive field enhancement in relaxor-based ferroelectric crystals
Limei Zheng, Liya Yang, Gang Liu
Shandong University, China.
- 14:30 B-4
Enhanced polarization property of PZT produced by press forming
Muneyasu Suzuki, Harutaka Mekar, Tetsuo Tsuchiya, Jun Akedo
AIST, Japan.
- 14:50 B-5
High Piezoelectric Performance of Textured Relaxor-PT/PZT Ceramics
Yan Yongke, Xu Zhuo, Priya Shashank
Xi'an Jiaotong University, China.
- 15:10 B-6
A Review of Lead Perovskite Piezoelectric Single Crystals and Their Medical Transducers Application
Yohachi (john) Yamashita, Tomoaki Karaki, and Horoshi Maiwa
Toyama Prefectural University, Japan.
- 15:30 B-7
Domain Engineering Effect of Alternating Current Poling with Different Frequency on PMN-PT Single Crystals
Chengtao Luo, Haotian Wan, Chang Liu, Wei-Yi Chang, Yohachi Yamashita, and Xiaoning Jiang
Shanghai Jiaotong University, China.

16:00-18:20 Invited Session C, Chair: Dou Zhang (Central South University)

16:00 C-1

Tuning of polar domain boundaries in non-polar perovskite

Nan Zhang, Zheyi An, Hiroko Yokota, Marek Paściak, Wei Ren, Zuo-Guang Ye

Xi'an Jiaotong University, China.

16:20 C-2

Microstructure Design and Exotic Electrostrain Response in Layer Structured Perovskite Ferroelectric Ceramics

Xiang He, Zhiguo Yi

Shanghai Institute of Ceramics, China.

16:40 C-3

Synergic Modulation of the Multi-scale Structures on the Energy Storage Properties of Silver Niobate-Based Ceramics

J. Wang, XH. Fan, Y. Rao, L. Zhao, KJ. Zhu

Nanjing University of Aeronautics and Astronautics, China.

17:00 C-4

Atomic-Scale Studies of PbZrO₃-based Antiferroelectric Ceramics

Zhengqian Fu, Xuefeng Chen, Zhenqin Li, Tengfei Hu, Ziyi Yu, Linlin Zhang, Shujun Zhang, Genshui Wang, Xianlin Dong, Fangfang Xu

Shanghai Institute of Ceramics, China.

17:20 C-5

Ferroelectric Materials and Devices for Intelligent Computing

Xiao Yu, Yue Peng, Jiuren Zhou, Genquan Han

Research Center for Intelligent Chips and Device, China.

17:40 C-6

Excellent pyroelectric properties of relaxor-based ferroelectric single crystals and thin films for infrared detection

Yanxue Tang, Haosu Luo

Shanghai Normal University, China.

18:00 C-7

Heterostructural magnetoelectric composites and application in magnetic field and current sensing

Jie Jiao, Rui Chen, Hanzhou Wu, Yaojin Wang, Haosu Luo, Li Lu, Wenning Di

Shanghai Institute of Ceramics, China.

Dec. 9, 2022 (Japan/Korea Standard Time)

9:00-11:00 Invited Session D, Chair: Jun Akedo, (AIST)

9:00 D-1

High Curie temperature BiFeO₃-BaTiO₃ lead-free piezoelectric ceramics: One-step preparation

Yu-Cheng Tang, Ai-Zhen Song, Yang Yin, He-Zhang Li, Bo-Ping Zhang
University of Science and Technology Beijing, China.

9:20 D-2

Bismuth Ferrite Based Lead-Free Piezoelectric ceramics

Dawei Wang

Harbin Institute of Technology, China.

9:40 D-3

Mechanism of Material Hardening on (Bi_{0.5}Na_{0.5})TiO₃-based Lead-free Piezoelectric Ceramics by Quenching Treatment

Takeru Tayama, Yuka Takagi, and Hajime Nagata

Tokyo University of Science, Japan.

10:00 D-4

Studies of Domain Structure in (K,Na)NbO₃-based Ceramics with Acid-Etching

Zhang Jialiang

Shandong University, China.

10:20 D-5

Effect of Li₂CO₃ and MnCO₃ Addition on Properties of (Ba,Ca)(Ti,Sn)O₃ Piezoelectric Ceramics Sintered Under Controlled Low Oxygen Partial Pressure

K. Uematsu, W. Sakamoto

Chubu University, Japan.

10:40 D-6

Investigations of multi-scale regulation and in-situ characterization of high temperature piezoelectric ceramics

Zhiyong Zhou

Shanghai Institute of Ceramics, China.

11:10-12:40 Poster Session, P-01—P-31

13:30-15:50 Invited Session E, Chair: Eisuke Tokumitsu (JAIST)

13:30 E-1

Enhanced energy storage ceramic capacitors utilizing ferrorestorable polarization

Yuji Noguchi, and Hiroki Matsuo

Kumamoto University, Japan.

13:50 E-2

High performance BaTiO₃-based dielectric materials for capacitor applications

Cheng Chen, Hua Hao, Xin Lai, Appiah Millicent, Zhonghua Yao, Minghe Cao, Hanxing Liu

Wuhan University of Technology, China.

14:10 E-3

Origin of High-Performance Energy Conversion in AgNbO₃ Based Ceramics: Structure Evidences

Zhigao Hu, Anyang Cui, Kai Dai, Kai Jiang, and Junhao Chu

East China Normal University, China.

14:30 E-4

Growth of Metal-free Perovskite-type Ferroelectric Crystals and Their Ferroelectric Properties

S. Ueno, S. Ando, T. Moriyama, T. Unno, I. Fujii, S. Kawachi, J. Yamaura, S. Wada

University of Yamanashi, Japan.

14:50 E-5

Effect of Reaction Accelerator on Single Crystal Barium Titanate Fine Particle Synthesis

Masaki YAMAGUCHI, Atsushi SAITOH, Misa YAMASAKI, Takashi YAMAMOTO

Shibaura Institute of Technology, Japan.

15:10 E-6

Ferroelectric and piezoelectric properties in simple binary compounds

Wei Ren

Shanghai University, China.

15:30 E-7

Angle-resolved polarized Raman mapping on a compositionally graded

Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃

S. Tsukada, Y. Fujii, A. Kanagawa, Y. Akishige, and K. Ohwada

Shimane University, Japan.

16:00-18:40 Invited Session F, Chair: Masaki Yamaguchi (Shibaura Institute of Technology)

16:00 F-1

Flexible Lead-free Piezoelectric Arrays for High-Efficiency Wireless Ultrasonic Energy Transfer and Communication

Laiming Jiang, Jiagang Wu, Jianguo Zhu

Sichuan University, China.

- 16:20 F-2
RTIC phenomenon on AD process and its application to dielectric / ferroelectric thick films
Jun AKEDO
National Institute of Advanced Industrial Science & Technology (AIST), Japan
- 16:40 F-3
Ferroelectric gate thin films transistors with Y-doped Hf-Zr-O gate insulator and In-Sn-O channel
E. Tokumitsu, Y. Kubota, Mohit, K. Sasaki
Japan Advanced Institute of Science and Technology, Japan.
- 17:00 F-4
Fabrication and characterization of La-doped HfO₂ ferroelectric films on SrTiO₃ single crystal substrates
Badari Narayana Rao, Hiroko Yokota
Chiba University, Japan.
- 17:20 F-5
Energy-storage performance of antiferroelectric Pb(Zr,Hf)O₃ films fabricated by sol-gel spin coating
J. Song, S. Kada, Y. Iwamoto, T. Iijima, and S. Okamura
Tokyo University of Science, Japan.
- 17:40 F-6
Low temperature deposition of BiFeO₃ films on base metal foils for piezoelectric applications
Jing Yan, Jun Ouyang, Isaku Kanno
Qilu Normal University, China.
- 18:00 F-7
Extraordinarily large domain switching contribution in epitaxial (K, Na)NbO₃ films
Xueyou Yuan, Kazuki Okamoto, Mitsuki Kawano, Masahito Yoshino, Takanori Nagasaki, Yasuhiko Imai, Osami Sakata, Tomoaki Yamada
Nagoya University, Japan.
- 18:20 F-8
Intrinsic properties of domain walls of BiFeO₃
Lisha Liu, Jing-Feng Li, and Yaojin Wang
Nanjing University of Science and Technology, China.
- 18:40 Closing remarks, Tomoaki Karaki (Zhiming Chen)

Poster Session

11:10-12:40, Dec. 9, 2022 (Japan/Korea Standard Time)

P-01

Phase Transformation, Interfacial Coupling and Giant Charge-discharge Efficiency of BaTiO₃@PVP/PVDF Nanocomposite Films

Yao Su, Wenyi Li, Yan Wang, Peimei Yuan, Zhao Bo, Dengwei Hu
Baoji University of Arts and Sciences, China.

P-02

Crystal structure and microwave dielectric properties of Ba₄(Sm_{1-x}Ce_x)_{9.33}Ti₁₈O₅₄ solid solutions

Yu Xie, Kongjun Zhu, Kang Yan, Jing Wang, Jinsong Liu
Nanjing University of Aeronautics and Astronautics, China.

P-03

Ultrahigh energy harvesting properties in temperature insensitive eco-friendly high performance KNN-based textured ceramics

Jinfeng Lin, Jiwei Zhai
Tongji University, China.

P-04

Characteristics of Chemical Bonding and Thermal Behavior of Atoms in Prototype Structure of Double Perovskite-type Antiferroelectric Oxides

Takayasu Shigemasu, Sangwook Kim, Chikako Moriyoshi, Guorong Li, Chul-Hong Park, and Yoshihiro Kuroiwa
Hiroshima University, Japan.

P-05

Tunable vertical ferroelectricity and domain walls by interlayer sliding in β -ZrI₂

Xiaonan Ma, Chang Liu, Sergey A. Nikolaev, and Wei Ren
Shanghai University, China.

P-06

Crystal Structure of KNbO₃/BaTiO₃ Core-Shell Nanocomposite Particles Studied by Synchrotron Radiation X-ray Diffraction

Shao Mingyang, Kaede Furuta, Sangwook Kim, Ichiro Fujii, Shintaro Ueno, Satoshi Wada, and Yoshihiro Kuroiwa
Hiroshima University, Japan.

P-07

Brillouin Scattering Study of Ferroelastic Phase Transition of Lead Phosphate Single Crystal

Adil Emin, Anwar Hushur, and Seiji Kojima
Lanzhou University, China.

P-08

Structural evolution and electrical properties of BFO-BT based high temperature lead-free piezoelectric ceramics

X. Ai, J. Yang, K. Yan, K. Zhu

Nanjing University of Aeronautics and Astronautics, China.

P-09

Activation of lead-free (Na,K)NbO₃ piezoceramics using pulsed electric field

Takumi Nozaki, Alexander Martin, Ryota Kobayashi, Tadachika Nakayama, Ken-ichi Kakimoto

Nagoya Institute of Technology, Japan.

P-10

Realization of enhanced strain responses in lead-free (Bi_{0.5}Na_{0.5})TiO₃-based ferroelectric thin films by active domain motions

Zhe Wanga, Jinyan Zhao, Gang Niu, Wei Ren, Nan Zhang, Kun Zheng, Yi Quan, Henghui Cai, Xin Li, Genshui Wang

Xi'an Jiaotong University, China.

P-11

Preparation of high temperature resistant barium titanate/polyimide flexible nanocomposite energy storage films

Peimei Yuan, Ruixuan Xue, ChenLi Wu, Rong Ma, Dengwei Hu

P-12

Change of crystal and microscopic structures of Ba_{0.95}Sr_{0.05}TiO₃ ceramics prepared by the combination of reaction sintering and spark plasma sintering

A Fujisawa, D Fu

Shizuoka University, Japan.

P-13

Microstructure of AC poled ternary PIN-PMN-PT single crystal after high temperature annealing

Yu Xiang, Yinqin Sun, Y. Yamashita, T. Karaki, and H. Maiwa

Shonan Institute of Technology, Japan.

P-14

Effects of sputtering conditions on the growth of AlScN thin films and its conduction mechanism

Kohei Miyaji, Norifumi, Fujimura, Takeshi Yoshimura

Osaka Metropolitan University, Japan.

P-15

High-Performance Neuromorphic Computing Based on Ferroelectric Synapses with Excellent Conductance Linearity and Symmetry

Shuting Yang, Limei Zheng, Le Zhao

Shandong University, China.

P-16

Low-temperature deposition of transparent $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ thin film on glass substrate with giant piezoelectric response

K. Yoshida, S. Sogen, H. Suzuki¹, T. Arai, and D. Fu
Shizuoka University, Japan.

P-17

Mechanomyography Using Piezoelectric Materials

S. Sakoda, N. Fujimura and, T. Yoshimura
Osaka Metropolitan University, Japan

P-18

Unique Gas Sensing Performance of Sol-gel Derived BiFeO_3 Nanoparticles

Xiaojie Li, Jinrong Cheng, Jiaqiang Xu
Shanghai University, China.

P-19

Ferroelectric and piezoelectric properties of c-axis oriented $\text{PbZr}_{0.53}\text{Ti}_{0.47}\text{O}_3$ thin film deposited on SUS substrate with large coefficient of thermal expansion

Seiji Sogen, Kazuto Yoshida, Takashi Arai, Hisao Suzuki, Desheng Fu
Shizuoka University, Japan.

P-20

Superior energy storage properties in NaNbO_3 -based ceramics via synergistically optimizing domain and band structures

Weiwei Yang, Huarong Zeng, Guorong Li
Shanghai Institute of Ceramics, China.

P-21

Extremely large strain response under low driving electric fields in lead-based textured piezoelectric ceramics

Hongrui Jia, Zhen Li, Linghang Wang
Xi'an Jiaotong University, China.

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Experimental approach to residual hydrogen content in silicon substrates by proton beam writing

Jun HIRADE and Masaki YAMAGUCHI
Shibaura Institute of Technology, Japan.

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Polarization Rotation Control Domain Dynamic Response Modulates Piezoelectric Properties of Lead-Free Thin Films

Kun Zhu, Guanglong Ge, Jinfeng Lin, Hairui Bai, Cheng Shi, Guohui Li, Fei Yan, Liuxue Xu, Weiwei Yang, Huarong Zeng, Kunyu Zhao, Zhenyong Man, Feifei Wang, Bo Shen, Jiwei Zhai, Xiujian Chou
Tongji University, China.

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High Energy Storage Performance and Large Electrocaloric Response in $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ Thin Films

Jin Qian, Jiwei Zhai, Zhenxiang Cheng

Tongji University, China.

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Enhanced stabilization of oxygen vacancies on the bipolar fatigue resistance of PNN-PZT-based ceramics

Ying Shi, Guorong Li, Liaoying Zheng, Zhenyong Man, Xuezheng Ruan, Xue Shi

Shanghai Institute of Ceramics, China.

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Preparation of Tetragonal BaTiO_3 by Molten Salt Synthesis Process

Fei Jing, Nina Dai, Weixing Zhao, Dengwei Hu

Baoji University of Arts and Sciences, China.

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Energy storage properties of antiferroelectric PbZrO_3 thin films prepared by chemical solution deposition method

T. Nishimoto, S. Sougen, K. Yoshida, H. Suzuki, D. Fu

Shizuoka University, Japan.

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3D Printing of Piezoelectric Biological Scaffolds for Bone Tissue Engineering

Ruixuan Xue, Peimei Yuan, Liying Yu, Rong Ma, Dengwei Hu

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Flexible Piezoelectric Nanogenerators Based on Two-dimensional Barium Titanate Nanosheets and Polymer Polylactic acid

Bo Zhao, Yao Su, Songlin Yu, Han Yu, Dengwei Hu

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Energy storage of BT/PVP/PVDF nanocomposite films with multilayer structure

Nina Dai, Fei Jing, Peimei Yuan, Ruixuan Xue, Bo Zhao, Yao Su, Dengwei Hu

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Fabrication of $(\text{Ba,Sr})\text{TiO}_3$ epitaxial thin films on mica substrate toward flexible electronics

Shicheng Peng, Sou Yasuhara, Takaaki Tsurumi, Takuya Hoshina

Tokyo Institute of Technology, Japan.

I-1:

Bragg Coherent X-ray diffraction for visualization of the inhomogeneous structure of a single ferroelectric particle

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The shapes and sizes of crystal grains affect the anisotropy of the crystal structure and the arrangement of ferroelectric domains [1,2]. These ferroelectric size effects, however, have made little progress in discussion due to the difficulty of simultaneously observing the crystal structure, external shape, and domain structure of a particle with sizes ranging from several 10 to several 100 nm.

We are developing and improving Bragg coherent diffraction imaging (Bragg-CDI) [3] at BL22XU at SPring-8[4], which is one of the promising methods to simultaneously visualize the heterogeneous structures (including domain structures) and external shapes of fine particles in a non-destructive manner. The Bragg-CDI system is now ready for application to ferroelectric size effect research since its applicable particle size has recently been expanded from the conventional 100-300 nm to 40-500 nm [5]. For application to single ferroelectric BaTiO₃ (BTO) particles with sizes of 200-500 nm, Bragg-CDI has visualized the reconstructed three-dimensional images with the external shape, size, and internal strain distribution of single particles. The 500-nm BTO particle showed a linear and sharp phase boundary shape, while smaller BTO particles showed different phase patterns.

To further understand these effects, we developed a method to track the ferroelectric phase transition in a BTO single particle [6]. The phase transition was observed by cooling the 500-nm-sized BTO to Curie temperature and recording the pattern changes in Bragg coherent X-ray diffraction. Cubic (200 peak)-to-tetragonal (200 and 002 peaks) transition was observed, with a bridge-like fringe pattern. Bragg-CDI also imaged the inhomogeneous structure of the BTO single particle in the cubic phase from the diffraction patterns. The reconstructed images showed linear strains due to dislocations inside the BTO particle.

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I-2:

Polar orders across scales: the effect of strain and mechanics

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Ferroelectrics are materials with long-range polar orders, and they have important applications in sensors, actuators, field effect transistors, and capacitors. Understanding polar orders across scales are thus of critical importance. In this talk, we will examine how ordinary domain patterns are accommodating each other across grains, resulting in extraordinary piezoelectric coefficient at morphotropic phase boundary. We will discuss how topologically nontrivial polar orders emerges and reconstruct in ferroelectrics, leading to exotic properties such as negative capacitance. We will also discuss how dielectric materials fail under high electric field, which has important implication for pulsed power capacitors. In all these examples, I will show how coupling to strain, and thus mechanics, play an essential role in the configuration and evolution of polar orders.

Energy Harvesting from Electric Power Lines Using Piezoelectric Resonator

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Energy harvesting is expected to be a promising power source for IoT devices. We have developed a dynamic model of piezoelectric vibration energy harvester (pVEH) and achieved a power density of $910 \mu\text{Wcm}^{-2}\text{G}^{-2}$ through device design based on this model and material development of BiFeO_3 thin film [1]. Moreover, using the knowledge obtained in this process, we have developed a harvester that uses permanent magnets and piezoelectric elements to extract electric power from the alternating magnetic field formed around a power line, which is called a magnetic field oscillation energy harvester (MOEH) [2]. A permanent magnet placed near a conductor with a current flowing through it is subjected to a force in a direction perpendicular to both the current and the magnetic moment. When an alternating current flows, the magnet oscillates, which can be used to generate electric power using the mechanism of pVEH.

The force F_x acting on a permanent magnet placed in a magnetic field with a gradient is given by

$$F_x = B_r \int \frac{dH_y(x, y)}{dx} dV \quad (1)$$

where B_r is the residual flux density of the magnet, V is the volume of the magnet, and H_y is the y component of the magnetic field. Based on the calculation of the force acting on the permanent magnet placed near the AC power line, the prototype of MOEH was fabricated. The resonant frequency of the harvester was adjusted to about 50 Hz. Fig. 1 shows the frequency dependence of the output power measured at various AC currents. The distance between the magnet and the power line was 12.2 mm. The output power at the resonant frequency increases in proportion to the square of the current and reaches to 0.35mW at an AC current of 3.4 A.

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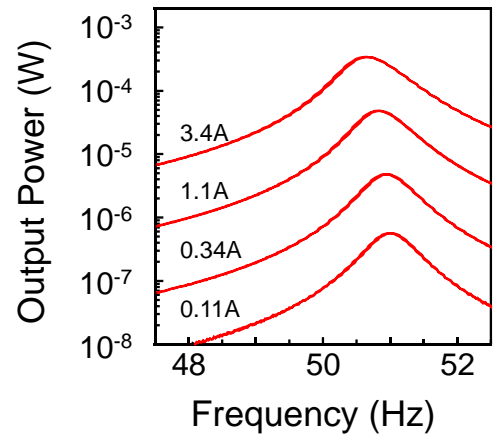


Fig. 1 frequency dependence of the output power measured at various AC currents.

A-2:

**Improved capacitive energy storage nanocomposites at high temperature
utilizing ultralow loading of bimetallic MOF**

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Abstract: It is urgent to develop high-temperature dielectrics with high energy density and high energy efficiency for next-generation capacitor demands. Metal organic frameworks (MOFs) have been widely used in the fields of electrochemistry, catalysis, gas storage, separation and sensing due to their structural diversity and functionally adaptable properties. Doping of metal nodes in MOFs is an effective strategy to change the band gap and band edge positions of the original MOFs, which helps to improve their ability to bind charges as traps. In this work, incorporation of ultralow loading (<1.5 wt%) of novel bimetallic metal-organic framework (ZIF 8-67) into PEI polymer matrix is exhibited. ZIF 8-67 is a kind of bimetallic frame material with excellent thermal stability, good insulation and microporous structure. With the addition of ZIF 8-67, the breakdown strength and energy storage capacity of ZIF 8-67/PEI nanocomposites are significantly improved, especially at high temperature (200 °C). For example, the energy densities of the 0.5 wt% ZIF 8-67/PEI nanocomposites is up to 2.96 J/cm³, with an efficiency (η) >90% at 150 °C. At 200 °C, the discharge energy density of 0.25 wt% ZIF 8-67/PEI nanocomposite can still reach 1.84 J/cm³ with a η >90%, which is 9 times higher than that of pure PEI (0.21 J/cm³) under the same conditions, and it is the largest improvement compared with the previous reports. Furthermore, at 200 °C, the energy storage density and efficiency of the nanocomposites show no sign of degradation after 20,000 cycles.

Ferroelectrics for emerging oral bioelectronics

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Tooth whitening has recently become one of the most popular aesthetic dentistry procedures. Beyond classic hydrogen peroxide-based whitening agents, photo-catalysts and piezo-catalysts have been demonstrated for non-destructive on-demand tooth whitening. However, their usage has been challenged due to the relatively limited physical stimuli of light irradiation and ultrasonic mechanical vibration. To address this challenge, we report here a non-destructive and convenient tooth whitening strategy based on the pyro-catalysis effect, realized via ubiquitous oral motion-induced temperature fluctuations. Degradation of organic dyes via pyro-catalysis is performed under cooling/heating cycling to simulate natural temperature fluctuations associated with intake and speech. Teeth stained by habitual beverages and flavorings can be notably whitened by the pyroelectric particles-embedded hydrogel under a small surrounding temperature fluctuation. Furthermore, the pyro-catalysis based tooth whitening procedure exhibits remarkably therapeutic biosafety and sustainability. In view of the exemplary demonstration, the most prevalent oral temperature fluctuation will enable the pyro-catalysis based tooth whitening strategy to have tremendous potential for practical applications.

High performance dielectric energy storage via nanograin engineering in ferroelectric/antiferroelectric film capacitors

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In comparison with the ferroelectric and antiferroelectric bulk ceramics, their thin films can achieve larger capacitive energy densities due to higher applicable electric fields. Recently, researchers have investigated different methods to improve the capacitive energy densities of ferroelectric or antiferroelectric ceramic film capacitors, such as “domain engineering”, “interface engineering” and chemical doping/substitution. In this work, we prepared high energy density film capacitors via a combination of the low-temperature sputtering process and the buffer-layer technique, as well as a rapid thermal annealing process. In comparison with the high-temperature processed films, our ferroelectric BaTiO₃ and CaBi₂Nb₂O₉ films and antiferroelectric Ag(Nb, Ta)O₃ films, displayed not only a greatly reduced thermal budget, but also a significantly decreased grain size (down to 10-20 nm). The latter has led to a slim and elongated P-E hysteresis loop and a high breakdown strength E_b, corresponding to a much improved energy storage density and a high charge-discharge efficiency. These low temperature processed films are also endowed with an excellent electric fatigue resistance and a high thermal stability. Applying phase field modeling on a simple linear model of the BaTiO₃ film consisting of periodically aligned and parallelly connected grains and grain boundaries, we explained its P-E characteristics and the corresponding capacitive energy storage performance (Fig.1).

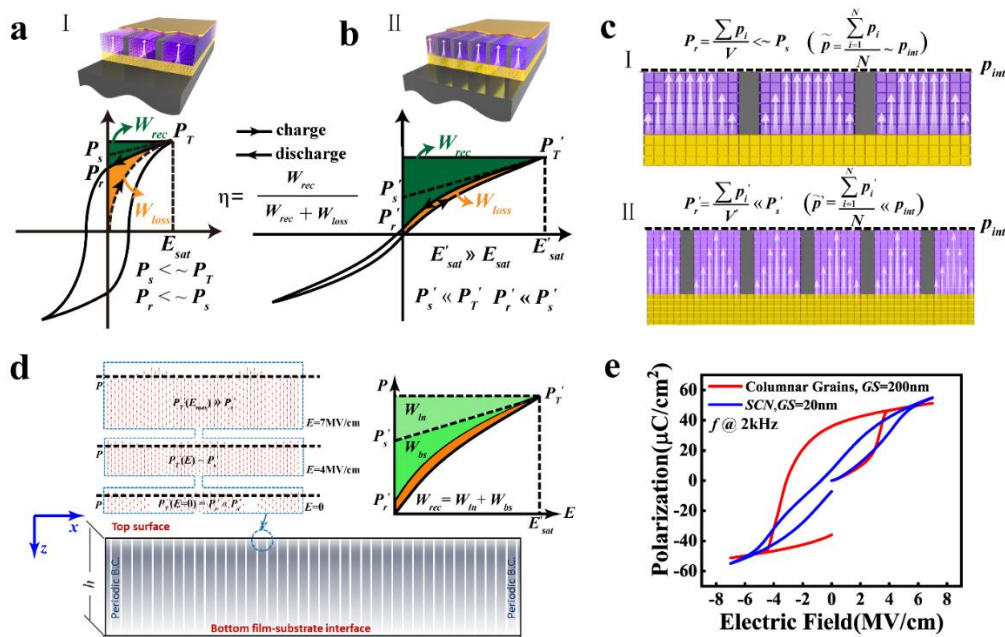


Fig1. Schematics of a high energy density columnar nanograin BaTiO₃ film, and its P-E characteristics simulated via phase field modeling (Zhao and Ouyang et al. *Energy Storage Materials*, **39**, 81, 2021).

Reducing the coercive field of rhombohedral NBT-based crystals by domain engineering

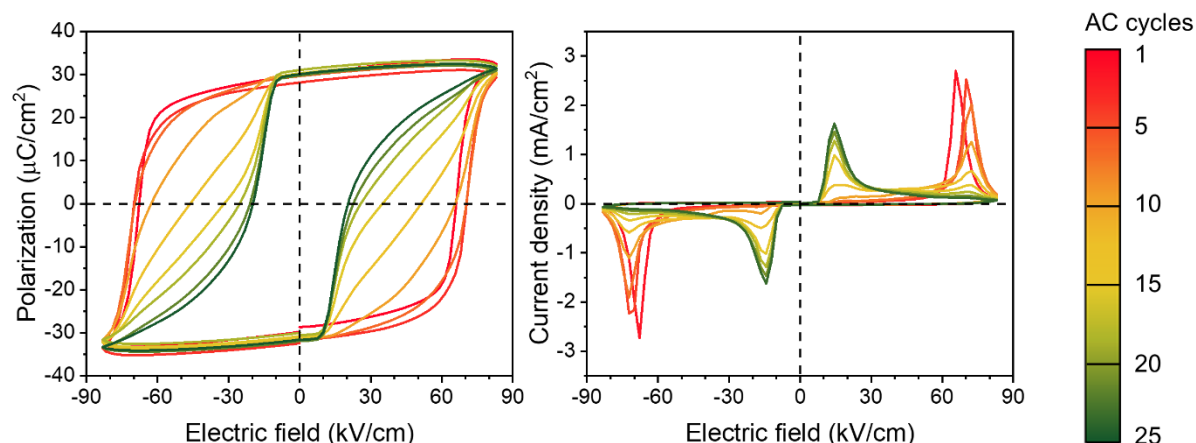
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($\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ (NBT)-based materials have been widely studied for their large electric-field-induced strains. However, the high driving voltage and coercive field (E_c) have long hindered the applications of NBT-based materials. Here, we propose a simple method to significantly reduce the E_c of rhombohedral NBT-based single crystals through tailoring the domain configuration. By applying an alternating-current (AC) electric field along the [001] direction, the E_c of Mn-doped ($\text{Na}_{0.48}\text{K}_{0.02}\text{Bi}_{0.5}\text{TiO}_3$) single crystal can be reduced from 70 kV/cm to 20 kV/cm after about 25 cycles without sacrificing the ferroelectric polarization. Meanwhile, the small-signal piezoelectric coefficient d_{33} and the optical transparency of the crystals are also improved compared with those after direct-current electric field poling.



The domain structure characterization shows that the AC cycles along the [001] direction can form a laminar domain configuration where the 71° domain walls are nearly eliminated, which is considered to be the cause of the reduced E_c . In the laminar domain configuration, almost only 71° polarization switching occurs when the external electric field is reversed, and 71° polarization switching has been proved to require lower energy than the 109° or 180° polarization switching in rhombohedral ferroelectric crystals. Therefore, the required energy for polarization reversal of the laminar domain configuration is significantly lower than that of the randomly distributed domains, thus the E_c is reduced greatly.

This method is also applicable to other rhombohedral single crystals (NBT, Mn-doped NBT, NBT-4%BaTiO₃, etc.) and may be applied to [001]-textured polycrystalline ceramics in the future to promote the practical applications of NBT-based materials.

Growth and Characterization of Ba₃TaGa₃Si₂O₁₄ Single CrystalsH. Takeda^{1,*}, T. Hoshina², and T. Tsurumi²¹Saitama University, 255 Shimo-Okubo, Sakura-ku, Saitama 338-8570, Japan²Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro, Tokyo 152-8552, Japan

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High temperature piezoelectric materials are in high demand for gas sensors, combustion pressure sensors and gas injectors directly placed in the cylinder of engines. La₃Ga₅SiO₁₄ (langasite; LGS)-type single crystals are currently promising materials for this purpose. The LGS crystal has Ca₃Ga₂Ge₄O₁₄-type structure (space group *P321*). There are four kinds of cation sites in the structure with the chemical formula *A*₃*BC*₃*D*₂O₁₄. The notations *A* and *B* represent a decahedral (twisted Thomson cube) site coordinated by eight oxygens, and an octahedral site coordinated by six oxygens, respectively. Moreover, *C* and *D* represent tetrahedral sites coordinated by four oxygens, the size of the *D* site being smaller than that of the *C* site. In the LGS crystal, the *A* site is occupied by La³⁺ ion, the *B* and *C* sites are occupied by Ga³⁺ ions, the *D* site is occupied by Ga³⁺ and Si⁴⁺ ions. Therefore, LGS is call as disordered-type. Due to the existence of these many cation sites, a large number of Ca₃Ga₂Ge₄O₁₄-type compounds have been synthesized by isovalent and/or aliovalent substitution [1]. Based on this data, Sr₃TaGa₃Si₂O₁₄ single crystals was synthesized. In this crystal, each cation site is occupied by only single metal [2]. Nowadays, this kind of crystals is call as ordered-type. It has been reported [3] that an increase lattice parameter a leads to an increase of the piezoelectric modulus $|d_{11}|$ of Ca₃Ga₂Ge₄O₁₄-type crystals. Based on these knowledge, Ba₃NbGa₃Si₂O₁₄ and Ba₃TaGa₃Si₂O₁₄ (abbreviated as BTGS) single crystals were synthesized and small information about their piezoelectric properties was disclosed [4,5]. As far as we know, there is no details of the structure analysis and piezoelectric properties of the BTGS single crystal.

We report structure and electric properties of the BTGS crystals grown by the Czochralski technique using raw materials with stoichiometric composition [6]. The crystal structure of BTGS has been refined using single-crystal X-ray diffraction data with a precision corresponding to an R index less than 0.04. The crystal structure is isostructural to that of langasite-type Ca₃Ga₂Ge₄O₁₄. It was confirmed that the distribution of each cation was ordered in each site. A piezoelectric d_{11} constant was 5.8 pC/N, which is the highest of those of order-type langasite crystals. The temperature dependence of resistivity is similar to those of order-type langasite crystals (Fig. 1) and higher than that of La₃Ta_{0.5}Ga_{5.5}O₁₄ (LTG) crystal, which is the most promising candidates for combustion pressure sensor materials.

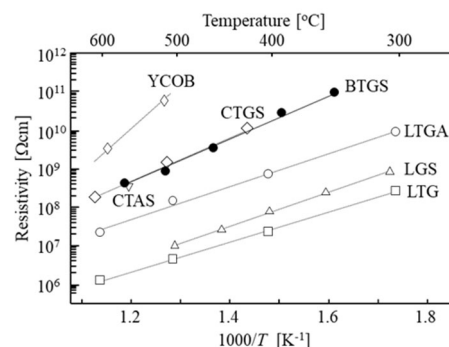


Fig. 1 Resistivity as a function of temperature for Ba₃TaGa₃Si₂O₁₄ (BTGS) crystals compared with other langasite family crystals. CTGS, CTAS, LTGA, LGS, and LTG are the abbreviations for Ca₃TaGa₃Si₂O₁₄, Ca₃TaAl₃Si₂O₁₄, La₃Ta_{0.5}Ga_{5.5}O₁₄, La₃Ga₅SiO₁₄, and La₃Ta_{0.5}Ga_{5.5}O₁₄, respectively.

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Effects of Al substitution on crystal structure and piezoelectric properties of $\text{Ca}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$ and $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ single crystals

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$\text{Ca}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$ [CNGS] and $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ [CTGS] piezoelectric single crystals with the langasite-type structure have been studied for applications of pressure sensor at high temperature and small oscillator with low power consumption because of their stable piezoelectric properties at high temperature, stable temperature coefficient of frequency and low impedance. Ga site in the CNGS and CTGS single crystals can be substituted by Al ion, and we reported the crystal growth and the piezoelectric properties of $\text{Ca}_3\text{Nb}(\text{Ga},\text{Al})_3\text{Si}_2\text{O}_{14}$ [CNGAS] single crystals with 0-50% Al concentrations and $\text{Ca}_3\text{Ta}(\text{Ga},\text{Al})_3\text{Si}_2\text{O}_{14}$ [CTGAS] single crystals with 0-100% Al concentrations^[1-4]. In the previous reports, the lattice parameters and piezoelectric properties systematically changed with the Al concentration. In this study, we conducted more detailed crystal structure analysis using X-ray to reveal the relationship between crystal structures and piezoelectric properties of CNGAS and CTGAS single crystals to clarify effects of Al substitution.

$\text{Ca}_3\text{Nb}(\text{Ga}_{1-x}\text{Al}_x)_3\text{Si}_2\text{O}_{14}$ [$x = 0, 0.25, 0.5$] and $\text{Ca}_3\text{Ta}(\text{Ga}_{1-y}\text{Al}_y)_3\text{Si}_2\text{O}_{14}$ [$y = 0, 0.25, 0.5, 0.75, 1.0$] single crystals were grown by the Czochralski method^[1,2] and crystal structures of single crystals were investigated by the X-ray diffractometer with an imaging plate. Bulk wave velocities were measured by a plane-wave ultrasonic material characterization system^[3,4]. Dielectric constants and coefficients of thermal expansion were determined by an impedance analyzer and a dilatometer, respectively.

Crystal structures of CNGAS and CTGAS single crystals could be obtained with high precision^[5], and Figure 1 is the SiO_4 tetrahedron and of the $\text{Ca}_3\text{Nb}(\text{Ga}_{0.5}\text{Al}_{0.5})_3\text{Si}_2\text{O}_{14}$ [$x = 0.5$] single crystal with bond lengths and angles. O(2)-Si-O(2) and Si-O(2)-(Ga,Al) bond angles systematically increased with the shift of Si in z-axis by increasing Al concentration. Cation shift of the Si ion out of center from the barycenter of the oxygen coordination environment in CNGAS and CTGAS single crystals systematically decreased with increasing Al concentration, and cation shift of the (Ga,Al) ion slightly decreased, suggesting the decrease of the dielectric constants are attributable to the decrease of cation shifts of Si and (Ga,Al) ions by Al substitution.

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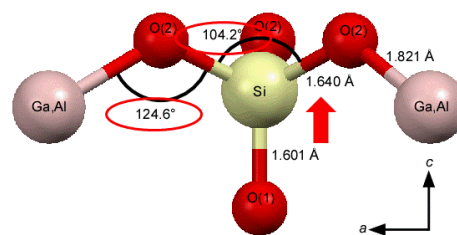


Fig. 1 Bond lengths and angles on SiO_4 tetrahedron of $\text{Ca}_3\text{Nb}(\text{Ga}_{0.5}\text{Al}_{0.5})_3\text{Si}_2\text{O}_{14}$ single crystal.

Simultaneously achieving giant piezoelectricity and record coercive field enhancement in relaxor-based ferroelectric crystals

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Both a large coercive field (E_C) and a giant piezoelectric activity are essential for the ferroelectric materials used in high-drive electromechanical applications, especially the case for greater power efficiency, smaller size, and lighter weight. A breakthrough in ferroelectric material is the discovery of relaxor-PbTiO₃ crystals, which have the highest piezoelectricity known today, but usually with low E_C being on the order of only 2-3 kV/cm. Such a performance deterioration is originated from the fact that the high piezoelectricity is usually interlinked to an easy polarization rotation, then favoring a dipole switch under small external stimuli. Therefore, searching for new types of ferroelectrics with simultaneously large E_C and ultrahigh piezoelectricity becomes an imminent challenge. Here, by adopting a microstructure design strategy involving small amount of scandium (Sc) doping, a novel ternary Pb(Sc_{1/2}Nb_{1/2})O₃-Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ crystal is reported, where the dispersed local heterogeneity featuring a considerable amount of tetragonal phase is confirmed. As a result, both elevated energy barriers and flattened potential wells are thermodynamically achieved, demonstrating a large E_C of 8.2 kV/cm (a factor of 3 greater than that of Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃) and ultrahigh piezoelectricity (d_{33} =2630 pC/N; d_{15} =490 pC/N). The observed E_C enhancement is the largest for ultrahigh-piezoelectric ferroelectric crystals, far exceeding the previously reported enhancements induced by the doping effects and other techniques. This updating investigation provides a simple, practical, and universal route for improving functionalities in ferroelectrics with an atomic-level understanding, which is critical for future material-by-design.

Enhanced polarization property of PZT produced by press forming

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Ferroelectrics are one of the most attractive materials to produce various electronic ceramics and to generate electricity, because of their high permittivity, excellent piezoelectricity and pyroelectricity derived from their unique crystal structures. In particular, lead zirconate titanate (PZT) is applied to various applications such as energy harvesting units, optical mirrors for laser displays, SAW/BAW filters, or gyrosensors, which are essential for supplying functionality or power to electrical devices. Since flexible substrates are sometimes indispensable in order to mount these oxide ferroelectrics on flexible or stretchable devices which have attracted attention recently, heat treatment in ceramic synthesis is often a problem. Therefore, advanced ceramics synthesis technique that eliminate for high-temperature process during manufacturing are strongly required in those research and development. In other words, low temperature synthesis of highly dense PZT ceramics that usually require sintering at temperatures above 1000°C via a solid-state reaction have been actively researched. For example, the PZT can be obtained at temperatures below 300°C by using cold sintering.¹⁾ Aerosol deposition allows the production of fully dense nanocrystalline ceramic films with a thickness of more than 1 μm at room temperature.²⁾ Technologies for transferring single-crystal thin films deposited by epitaxial growth or high-temperature crystallized films onto plastic substrates have also been advanced.³⁾ Thin films have been successfully formed by using regularly arrayed nanomaterials such as nanocubes.⁴⁾

We developed a new press forming-based, room temperature process, the mega-press forming (MF) method, that produces highly dense aggregate oxide films at a pressure below 1 GPa, which is lower than that at which oxide particles fracture, by taking advantage of the differences in the cohesive force between microsized and nanosized particles.⁴⁾ The MF method allowed us to produce high dense ceramic films with both high gas permeability and high impregnability. We succeeded in producing a PZT MF film with relative density of 94 % on an aluminum foil at room temperature that when impregnated with silicone oil exhibited saturated polarization hysteresis [see fig.1].

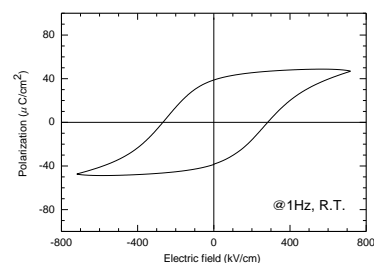


Fig.1 Polarization property for PZT film produced by press forming at room temperature on Al foil. This PZT film was not done any heat treatment.

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High Piezoelectric Performance of Textured Relaxor-PT/PZT Ceramics

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High performance piezoelectric materials have attracted great attention from both the high-tech application and fundamental research due to wide applications as actuators, sensors, and transducers. Particularly, textured piezoelectric ceramics are highly demanded, considering of excellent performance similar to that of single crystal and low cost similar to that of random ceramics. In this talk, we first present our prior achievements in textured relaxor-PT/PZT ceramics, and then introduce our recent progress on achieving ultra-high piezoelectricity in rare-earth doped textured PMN-PT ceramics and near-ideal electromechanical coupling in highly textured PMN-PZT ceramics. Several representative characteristics of textured piezoelectric ceramics will be highlighted:

High energy density: We demonstrated textured PMN-PZT ceramics which provide highest known energy density in piezoelectric materials through simultaneously enhancement of piezoelectric charge and voltage coefficients (d and g). We obtained a giant magnitude of $d \cdot g$ coefficient with value of $59000 \times 10^{-15} \text{ m}^2 \text{ N}^{-1}$ (comparable to that of the single crystal counterpart and 360% higher than that of the best commercial compositions).

High piezoelectric voltage coefficient: Single-phase oxide piezoelectric materials with giant piezoelectric voltage coefficient (g , induced voltage under applied stress) and high Curie temperature (T_c) are crucial towards providing desired performance for sensing, especially, under harsh environmental conditions. We provided a rational design of high g textured piezoelectric ceramics for sensing via incorporating (a) anisotropy/composition/phase structure, (b) microstructure and (c) domain engineering. We reported a $\langle 001 \rangle$ textured PbTiO_3 -based ceramics that has a high T_c ($\sim 364^\circ \text{C}$) and an extremely large g_{33} ($115 \times 10^{-3} \text{ Vm/N}$) in comparison to other known single-phase oxide materials.

Near-ideal electromechanical coupling: Electromechanical coupling factor, k , of piezoelectric materials determines the conversion efficiency of mechanical to electrical energy or electrical to mechanical energy. We reported a near-ideal k value of 0.93 in textured PMN-PZT which reaches the same magnitude as that for a single crystal, far beyond the average value of traditional ceramics. This demonstration of near-ideal k value in textured ceramics will have tremendous impact on design of ultra-wide bandwidth, high efficiency, high power density, and high stability piezoelectric devices.

B-6:

A Review of Lead Perovskite Piezoelectric Single Crystals and Their Medical Transducers Application

Yohachi (john) Yamashita^{1,2,3}, ¹Tomoaki Karaki, and ²Horoshi Maiwa

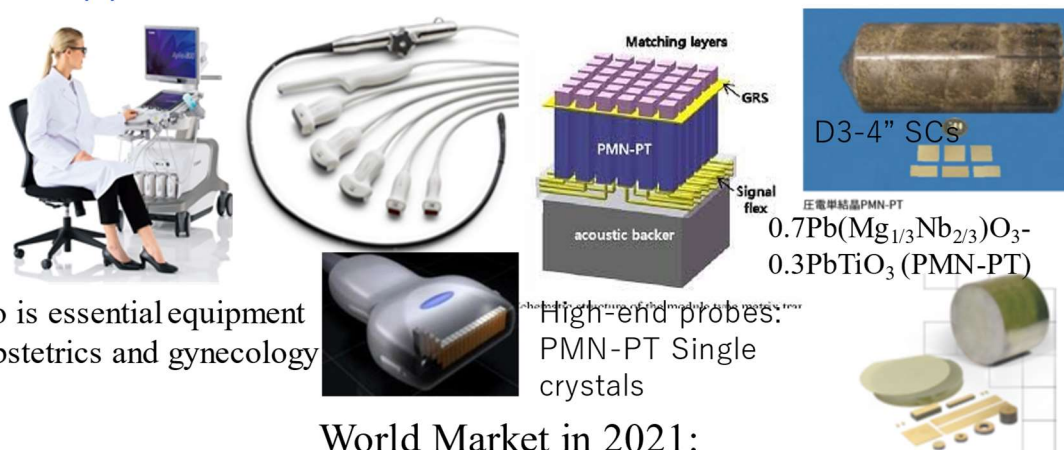
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Piezoelectric materials have been developed since early 1900s and many research had been conducted on the composition and process to obtain higher piezoelectric constants (d_{33}). Within composition research, lead perovskite relaxor piezoelectric single crystals (SCs) of binary $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - lead titanate PbTiO_3 (PMN-PT) and ternary $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3$ -PMN-PT SCs have been actively studied since 1990s because of their outstanding $d_{33} > 1,500$ pC/N compared to those of conventional $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ ceramics. A major driving force of these SC research has been promoted by mass-production of ultrasound transducers and arrays probes for medical diagnostic systems since early 2000s. However, the higher d_{33} material and process research for these ultrasound devices are almost saturated. In this review article, we present a brief overview of the history, current situation, and future perspective of piezoelectric SCs. Authors believe that main research in the next century is high d_{33} SCs with a high composition uniformity and low-energy SC growth methods, such as solid-state-SC growth, low-material loss SC transducer manufacturing technique, and improved poling process. This is a big technical challenge for all scientists; however, the relatively large market of medical ultrasound has been expanded year by year, and we hope that the community is motivated to solve such technical problems in the near future.

Application of relaxor-PT SCs: Medical Echo

9



Echo is essential equipment in obstetrics and gynecology

High-end probes: PMN-PT Single crystals

World Market in 2021:

Echo system \$6 B.US, Probes: \$0.6 B. US, SCs: \$40-50 M.US.

>1.0 M. SC probes have manufactured in last 2 decades

Domain Engineering Effect of Alternating Current Poling with Different Frequency on PMN-PT Single Crystals

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The alternating current (electric field) poling (ACP) was applied on [001]-oriented 0.7Pb(Mg_{1/3}Nb_{2/3})O₃ - 0.3PbTiO₃ (PMN-0.3PT) single crystal samples with dimensions of 5-mm-thick k33 mode and the influence of ACP frequency (fACP) was studied. Compared to those from traditional direct (electric field) poling (DCP) samples, d₃₃ and $\epsilon T_{33}/\epsilon_0$ of ACP samples could gain up to 67% increase to 3200 pC/N and 10500, respectively. In this work, low fACP is preferred for better enhancement. The ACP dynamics and the corresponding domain engineering effect during the saturation process were studied by both P-E loops and phase-field simulation.

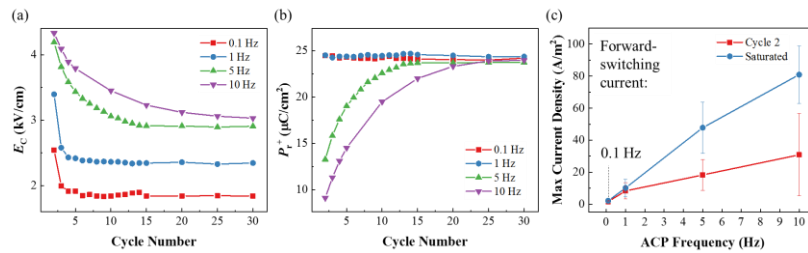


Figure 1. The saturation process as a function of cycle number (cycle 2 to saturated cycle 30) for (a) coercive field and (b) positive remnant polarization (c) The current density changes between unsaturated (cycle 2) and saturated cycles are compared.

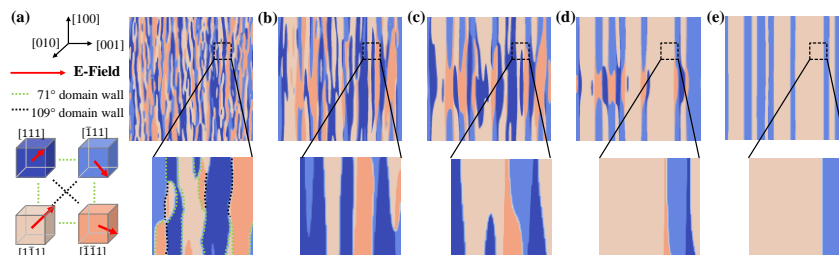


Figure 2. Phase-field simulation of the domain structures evolution during ACP from '4R' to '2R' configuration.

Tuning of polar domain boundaries in non-polar perovskite

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Domain-boundary engineering possesses great potential in nanotechnology industries. Domain boundaries in ferroic materials, as 2D topological defects, are found to have various physical properties not found in the surrounding domains, such as high conductivity, photovoltaic effect, and chirality. These properties can be enhanced and bring promising functionalities when centrosymmetric non-polar materials encounter polar domain boundaries.

In this work, a tunable polar domain boundary is discovered in an antiferroelectric single crystal. Under a small stress or electric field, the density, volume, and polarity of the boundaries are successfully controlled. We first confirmed the macroscopic polarity of the domain boundaries in antiferroelectric $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ (PZT) single crystals, using Second Harmonic Generation (SHG). Structural studies unveiled that the origin of the polarity comes from the atomistic structure of the antiphase boundaries, which possess a slightly different ionic displacement pattern from the bulk. Because of the structural compatibility, they occur more abundantly than in other ferroelastic materials and are proved to be easier to control. The domain boundaries can be preferentially created and erased by applying a small stress. The polarity of domain boundaries can be enhanced or switched with a small electric field. This work sheds light on this area not only by confirming its potential application in nanotechnology but also by promoting a fundamental understanding on this issue.

Microstructure Design and Exotic Electrostrain Response in Layer Structured Perovskite Ferroelectric Ceramics

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It is well known that the electrostrain of ferroelectric materials mainly comes from the contributions of intrinsic strain (electrostriction and piezoelectric response) and extrinsic strain (domain inversion and phase transition). In the past few decades, in order to improve the electrostrain output of ferroelectric materials, researchers have generally used crystal structure regulation and Morphotropic phase boundary design to improve the electromechanical response characteristics of samples, and achieved great success in lead containing material systems. Its typical feature is the significantly enhanced butterfly strain curve value. Compared with lead containing materials, Bi_2WO_6 has the advantages of high Curie temperature and environmental friendliness, and is a promising high-temperature piezoelectric actuator material. However, due to the two-dimensional lamellar grain growth habit, large polarization anisotropy and low piezoelectric activity of Bi_2WO_6 , its electromechanical response characteristics have not been reported. In this paper, bismuth layered Bi_2WO_6 ferroelectric ceramics with different microstructures were prepared by combining discharge plasma sintering and traditional solid phase sintering. A strange loop shaped strain curve was observed in textured Bi_2WO_6 ceramics for the first time. The maximum apparent strain was up to 0.75%, and it also had good cycle stability, superior to most lead-free ferroelectrics. It is worth noting that this loop type strain curve is different from the asymmetric strain electric field curve observed in traditional hard doped ceramic materials, and its asymmetric direction does not reverse with the sample turnover. Through repeated verification of the test system and test conditions, combined with piezoelectric response force microscope observation and X-ray photoelectron spectroscopy analysis, the author revealed that the singular strain curve originated from the reversible bending deformation caused by the asymmetric inversion of 90° ferroelastic domains on the upper and lower surfaces of ceramic materials. Unlike the electromechanical response characterized by butterfly loops, which reflects the uniform expansion or contraction of the sample along the thickness direction under the action of electric field, and also different from the asymmetric strain curve caused by defects or internal bias fields, this study reveals a new strain mode that may exist in ferroelectric materials, that is, the bending deformation caused by the non-uniform deformation of the sample under the action of electric field. This discovery not only provides a new idea for the design of piezoelectric actuators, but also provides a new idea for the origin of asymmetric strain curves.

C-3:

Synergic Modulation of the Multi-scale Structures on the Energy Storage Properties of Silver Niobate-Based Ceramics

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Silver niobate AgNbO₃ ceramics have been regarded as a promising lead-free material for energy storage applications. Benefiting from the hydrothermal synthesised AgNbO₃ powders, AgNbO₃ ceramics with fine-grain of ~3.4 μm were obtained. The fine-grain leads to increased electric breakdown strength E_b up to 250 kV/cm, which is the highest among the pure AgNbO₃ ceramics to our best knowledge. Extending beyond prior art for the compositional modification, synergic modulation of the multi-scale structures on the energy storage properties of AgNbO₃-based ceramics was reported, in which the introduction of MnO₂ at the atomic-scale improves the electric field of the FE-AFE phase transitions (E_A) while the SiO₂ coating on the AgNbO₃ particles at the micro-scale greatly increases the electric field inducing the AFE-FE transitions (E_F) and E_b . As a result, 0.10 wt% MnO₂-doped AgNbO₃@SiO₂ ceramics reveal a recoverable energy density up to 3.34 J/cm³ and efficiency of 60.0%, indicating 2.3 and 1.6 times larger than its counterpart of pristine AgNbO₃. Furthermore, equivalent amount Bi₂O₃ and Sc₂O₃ were subsequently introduced to improve the energy storage property of 0.10 wt% MnO₂-doped AgNbO₃@SiO₂ ceramics by simultaneously enhancing the maximum polarization, breakdown strength and relaxation feature. Of particular interest is that the AgNbO₃-based ceramics with 4 mol% Bi₂O₃ and Sc₂O₃ demonstrate the recoverable energy storage density of 5.9 J/cm³ with the energy storage efficiency of 71%. In addition, the benign energy storage performance can be maintained at elevated temperatures, frequencies and up to 10⁵ cycling, foreshowing great potential in advanced high-power applications. Our work provides a rational approach to synergically modulate the energy storage properties via the design of multi-scale structures.

Atomic-Scale Studies of PbZrO₃-based Antiferroelectric Ceramics

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Abstract:

Benefitting from the reversible phase transition between antiferroelectric and ferroelectric states, antiferroelectric materials have recently received widespread attentions for energy storage applications. Antiferroelectric configuration with specific antiparallel dipoles has been used to establish antiferroelectric theories and understand its characteristic behaviors. In this presentation, we present that the so-called antiferroelectric (Pb,La)(Zr,Sn,Ti)O₃ system is actually ferrielectric in nature. We demonstrate that the width and net polarization of ferroelectric ordering segments can be tailored by composition, which is linearly related to the key electrical characteristics, including switching field, remanent polarization and dielectric constant. Furthermore, we show how the modulated structure respond to external stimulus. These findings provide opportunities for comprehending structure-property correlation and fundamental understanding on antiferroelectric materials.

Ferroelectric Materials and Devices for Intelligent Computing

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Recently, transistor scaling is approaching its physical limit, hindering the further development of the computing capability, as shown in Fig. 1(a). The first is “power wall”, which block the enhancement of main frequency of the processor due to rising power density and heating effect; the second is “memory wall”, which is induced by huge performance or area gap between logic device and memory cell. The last is called Von Neumann bottleneck (BN) that huge data move between memory and processor would induce obvious delay and power issues.

In the post-Moore era, emerging logic and storage devices have been the fundamental hardware for expanding the capability of intelligent computing. Among them the emerging ferroelectric materials-based devices could reduce the obstacles of “power wall” “memory wall” and Von Neumann bottleneck, as illustrated in Fig. 1(b). In this presentation, the recent progress of ferroelectric devices for intelligent computing is discussed. The material properties and electrical characteristics of ferroelectric devices are elucidated, followed by a discussion of novel ferroelectric materials and devices that can be used for intelligent computing. Ferroelectric capacitors, transistors, and tunneling junction devices used for low-power logic, high-performance memory, and neuromorphic applications are comprehensively reviewed and compared. In addition, to provide useful guidance for developing high-performance ferroelectric-based intelligent computing systems, the key challenges for realizing ultrascaled ferroelectric devices for high-efficiency computing are discussed.

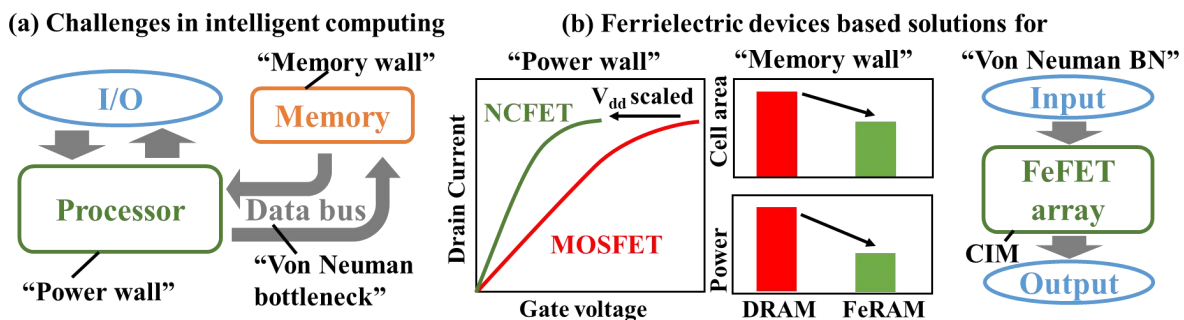


Fig. 1 Schematics of (a) challenges faced by modern computers using von Neumann architecture and (b) solutions for “heat wall”, “memory wall”, and von Neumann bottleneck based on ferroelectric devices.

Excellent pyroelectric properties of relaxor-based ferroelectric single crystals and thin films for infrared detection

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Infrared detectors and imagers based on a pyroelectric detection mechanism can be operated at ambient temperatures without cooling systems, as well as having broader spectral responsivity compared to the photon-based ones. The development of highly-sensitive, high-stability, and low-power uncooled infrared detectors and imagers has a strong need for pyroelectric materials with high pyroelectric coefficients and low dielectric loss. All pyroelectric crystals are also piezoelectric, the thermal expansion induced by heating leads to a strain which operates through the piezoelectric effect to give an additional secondary pyroelectric effect, therefore, large primary but low secondary pyroelectric coefficients are desirable for pyroelectric sensors.

We have investigated the composition, orientation, and temperature dependence of structures and pyroelectric properties for pure and Mn-doped binary $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ and ternary $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ relaxor-based ferroelectric single crystals and thin films. The secondary pyroelectric coefficients were calculated using thermodynamics method. Excellent pyroelectric properties with good thermal stability have been achieved for infrared detecting and imaging applications. The essential relationship between pyroelectric properties and microstructures have been determined. The high figures of merit, together with good thermal stability, make the materials promising candidates for high-performance infrared detectors and thermal imagers.

Key words: Ferroelectric, Pyroelectric properties, Infrared detection

Heterostructural magnetoelectric composites and application in magnetic field and current sensing

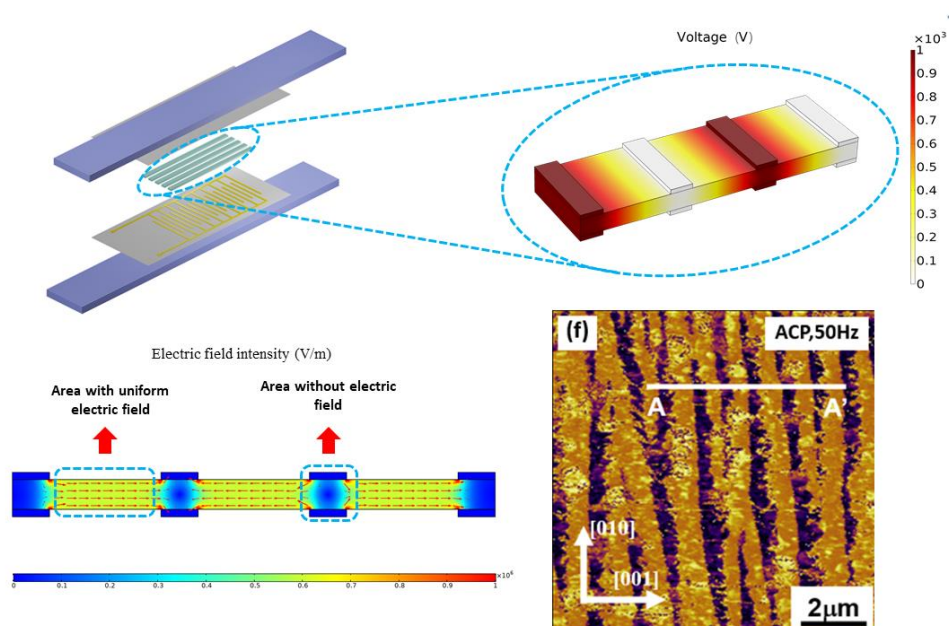
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Abstract: High-performance weak magnetic sensors have important uses in medical diagnostics, resource exploration, military anti-submarine and automotive electronics. In this study, we optimize the overall performance of magnetoelectric weak magnetic sensors from the perspective of noise-equivalent magnetic field by both reducing the noise level and improving the magnetoelectric response, and explore its application prospects in the field of current sensing. Magnetoelectric composites are the core of magnetoelectric sensors, whose noise at low frequencies mainly originates from DC resistance noise and dielectric loss noise, of which dielectric loss noise dominates. It was found that annealing under high-temperature oxygen-rich conditions significantly reduces the dielectric loss of the crystal, resulting in a reduction in the overall noise level of the sensor. On the other hand, the piezoelectric performance is enhanced by using AC polarization to enhance the non-intrinsic piezoelectric effect, resulting in a 54.6% increase in the responsivity of the magnetoelectric sensor at low frequencies. Based on the optimization results of the above two aspects, the noise equivalent magnetic field of the magnetoelectric sensor prepared by exploiting the bulk effect reaches 0.7 ± 0.2 pT/Hz^{1/2}@1Hz, and the detection capability far exceeds that of the embargoed version of the fluxgate (6 pT/Hz^{1/2}@1Hz). Compared to conventional current sensors (current transformers, Hall effect current sensors, etc.), magnetoelectric devices are considered as a new solution for current sensing due to their low power consumption and high sensitivity. Here, we report a current visualization sensor, working on the magnetoelectric/electrochromic principle, that converts the current energy in the environment into an electrical signal and subsequently into an easily observable color change, which has important applications in current monitoring, alarming, etc.



D-1:

High Curie temperature BiFeO₃-BaTiO₃ lead-free piezoelectric ceramics: One-step preparation

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A notorious problem in BiFeO₃-based piezoelectric ceramics is how to limit the formation of the impurities Bi₂₅FeO₃₉ and/or Bi₂Fe₄O₉ to give them excellent piezoelectric performance. Based on the Gibbs free energies of BiFeO₃, Bi₂₅FeO₄₀ and Bi₂Fe₄O₉, a new one-step calcining and sintering process was proposed to prepare BiFeO₃-*x*BaTiO₃ (BF-*x*BT) ceramics at a low sintering temperature, which can prevent impurity formation and realize the low-temperature sintering of single BF-*x*BT ceramics. The resultant ceramics are compact, porous-less, display macro-scale stripe domains along with a small amount of island-like domains, and exhibit saturated *P-E* loops, typical *I-V* characteristics, butterfly *S-E* curves and excellent piezoelectric properties ($d_{33} = 198\text{--}203$ pC/N; $T_C = 494\text{--}513$ °C). A careful analysis of the XRD patterns of BF-*x*BT ($0 \leq x \leq 1$) powder at different calcination temperatures reveals that the diffusion reaction path of ions is critical to prevent the formation of the impurities. It is also found that the Bi content and sintering temperature (T_{sint}) have a little effect on the phase structure, insulation, ferroelectric, piezoelectric and dielectric properties for 0.7Bi_(1+x)FeO₃-0.7BaTiO₃ ceramics prepared by one-step preparation. Stable piezoelectric properties of $d_{33} = 180\text{--}205$ pC/N, $P_r = 17.9\text{--}23.8$ μC/cm², $P_m = 25.9\text{--}33.6$ μC/cm², $T_C = 485\text{--}518$ °C was achieved at $-0.03 \leq x \leq 0.05$ and 930 °C $\leq T_{\text{sint}} \leq 970$ °C. Our results clarify the reaction mechanism and provide a synthesis strategy for designing high performance piezoelectric materials with good stability and easy industrialization.

Bismuth Ferrite Based Lead-Free Piezoelectric ceramics

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Bismuth ferrite (BF)-based solid solutions are considered candidates as lead-free functional ceramics due to their promising ferroelectric properties. Among various BF-based solid solutions, $(1-x)\text{BiFeO}_3-x\text{BaTiO}_3$ (BF-BT) has attracted attention for compositions within or close to a mixed phase region (MPR) in which rhombohedral and cubic polymorphs coexist. These compositions have high ferroelectric Curie temperature ($T_C > 400$ °C) but are also reported to have large remanent (P_r) and saturation (P_s) polarization coupled with large piezoelectric coefficients (d_{33}), particularly for samples quenched from the sintering temperature. More importantly, large electric field-induced strains and strain coefficients have been extensively reported in doped BF-BT within the MPR for $0.25 < \text{BT} < 0.35$. The element doping or substitution of a third component (often a nominal perovskite composition) in the BF-BT solid solution, results in a ferroelectric to relaxor-ferroelectric phase transition and further enhances strain. In this presentation, recent progress on the high electrostrains for BF-BT-based lead-free piezoelectric ceramics will be reviewed and the origin of the large electrostrain using *in situ* poling synchrotron X-ray diffraction will be discussed.

Mechanism of Material Hardening on (Bi_{0.5}Na_{0.5})TiO₃-based Lead-free Piezoelectric Ceramics by Quenching Treatment

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Bismuth sodium titanate (Bi_{0.5}Na_{0.5})TiO₃ (BNT)-based solid solutions have relatively high piezoelectric properties and mechanical quality factor in lead-free piezoelectric materials; therefore, they have attracted attention as the candidate lead-free materials for ultrasonic devices. However, BNT has the problem of having a low depolarization temperature T_d of approximately 180°C, resulting in narrow working temperature. In particular, the morphotropic phase boundaries (MPBs) of BNT-based solid solutions have a low T_d of approximately 100°C, even though an MPB has excellent piezoelectric properties. That is, the piezoelectric constant and T_d show a trade-off relationship, so that the elevation of T_d without the deterioration of piezoelectric activities is strongly demanded for practical piezoelectric applications using BNT-based ceramics.

Recently, our group has reported that a quenching procedure during the sintering process was an effective way of increasing the T_d of pure BNT ceramics without deteriorating their ferroelectric and piezoelectric properties.¹⁾ The T_d of a BNT sample quenched from 1100 °C was 223 °C, which was almost 50 °C higher than that prepared by the ordinary cooling process. On the other hand, it is generally known that quenching process affects the mechanical bending strength of the ceramics weakened by a thermal shock. Since the T_d increased by the quenching treatment from above 800°C in our previous study, we controlled quenching rate QR in the range from 1100°C to 800°C. Then, both T_d and mechanical strength reached higher values by controlling the QR.

In the presentation therefore, we will focus on a mechanism of the quenching effects and its influence for electrical and mechanical properties on BNT-based ceramics. Then, it is concluded that controlling QR is an effective method to improve both T_d , piezoelectric properties and mechanical strength simultaneously. Furthermore, we have applied the quenching procedure to the BNT-based solid solution systems with MPB and tetragonal phase. Even though in the MPB and tetragonal phases, BNT-based solid solution system (BNT-BaTiO₃ system) indicated higher T_d after the quenching treatment.²⁾ Piezoelectric performances were slightly decreased, whereas the T_d was increased significantly. Additionally, an elastic compliance s_{33}^E was decreased and a mechanical quality factor Q_m was increased after the quenching treatment. In other words, a hardening effect was observed after the quenching treatment on this composition. We will also discuss about the reasons of hardening effects by quenching treatment from the measurements of oxygen vacancies (by SIMS) and microstructure (by SEM and TEM). Then, a product of piezoelectric constant d and Q_m (dQ product: one of the figure of merit for high-power piezoelectric properties) was increased after the quenching treatment. We think this is good sign for the high-power piezoelectric applications.

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D-4:

Studies of Domain Structure in (K,Na)NbO₃-based Ceramics with Acid-Etching

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Piezoelectric performance of a ferroelectric ceramic can be greatly affected by its domain structure, which in turn is usually determined by chemical composition, microstructure, phase state and temperature. How to characterize the domain structure is important for the understanding of piezoelectric performance. The structural observation of thin specimens with a transmission electron microscope (TEM) and the morphological observation of chemically etched surface with an optical microscope or a scanning electron microscope (SEM) are the two commonly used methods.

The TEM observation can give the details of both lattice structure and nanodomain structure owing to high magnification. However, it requires very thin specimens that is sensitive to the preparation procedure, in which domain structure may change irreversibly due to the mechanical stresses of grinding, dimpling and focused ion-beam milling. Further, grains in a thin ceramic specimen are obviously in the different clamping state from those in a bulk ceramic, which may also lead to a substantial change in the domain structure. The case becomes even more critical when observing the domain structure for those ferroelectric ceramics that have one phase transition occurring around room temperature. Additionally, the obtained information comes only from a very limited area and thus may not be representative, but be regional with large randomness since the actual domain structure may differ significantly from grains to grains. Therefore, it is safe to take great caution when reading those results obtained by the TEM observation. On the other hand, acid-etching is considered as a convenient and quite reliable technique. Its working principle is based on the difference in etching rate between two adjacent domains of different polarization orientations at the specimen surface. Since the etched domain patterns can be easily observed with an optical microscope or a scanning electron microscope (SEM), acid-etching facilitates the study of domain structure for a large number of grains and makes it possible to acquire the overall characteristic and regularity, thereby enabling us to draw the credible conclusions on the basis of statistical information.

Due to the reasons described above, the acid-etching method has been frequently adopted in our studies of the various KNN-based ceramics. This talk will mainly report the results obtained in our two recent studies. One is to explore the compositional effects on piezoelectric properties, phase transitions and domain structure in a series of $(1-x)(\text{K}_{0.48}\text{Na}_{0.52})\text{Nb}_{0.955}\text{Sb}_{0.045}\text{O}_3-x(\text{Bi}_{0.50}\text{Na}_{0.50})\text{HfO}_3$ ceramics with $0.01 \leq x \leq 0.04$. The other is to systematically investigate the evolution of domain structure in the $0.96(\text{K}_{0.48}\text{Na}_{0.52})(\text{Nb}_{0.96}\text{Sb}_{0.04})\text{O}_3-0.04(\text{Bi}_{0.50}\text{Na}_{0.50})\text{ZrO}_3$ ceramics of a large d_{33} of 512 pC/N with poling and temperature.

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Effect of Li_2CO_3 and MnCO_3 Addition on Properties of $(\text{Ba,Ca})(\text{Ti,Sn})\text{O}_3$ Piezoelectric Ceramics Sintered Under Controlled Low Oxygen Partial Pressure

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$\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT)-based materials have been widely used as piezoelectric ceramics for a long time. However, these materials contain toxic PbO as a major component. Because of growing concerns about the global environmental issues, development of materials which do not contain toxic elements is strongly demanded. Among several lead-free piezoelectric ceramics, BaTiO_3 -based materials have been receiving much attention as a potential candidate. Chemical composition modification of BaTiO_3 has been extensively investigated to realize the desired electrical properties. Furthermore, for the development of the multilayer devices with high performance and low cost, reduction-resistant piezoelectric ceramics, which can use base metal electrodes, are also important.

In this study, reduction-resistant lead-free $(\text{Ba,Ca})(\text{Ti,Sn})\text{O}_3$ piezoceramics with high piezoelectric constants were firstly investigated by optimizing the Li_2CO_3 addition. For the sintering of the $(\text{Ba,Ca})(\text{Ti,Sn})\text{O}_3$ ceramics in a reducing atmosphere, the oxygen partial pressure was controlled by using $\text{Ar-H}_2\text{-CO}_2$ mixed gas. Enhanced grain growth and a high polarization state after poling treatment were achieved by adding Li_2CO_3 to $(\text{Ba,Ca})(\text{Ti,Sn})\text{O}_3$. Among various amount of Li_2CO_3 added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$, $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ ceramics with 3 mol% Li_2CO_3 addition were found to exhibit excellent piezoelectric properties while maintaining the high sintered density. Sintered bodies of the 3 mol% Li_2CO_3 -added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ ceramics prepared in a $\text{CO}_2(1.5\%)\text{-H}_2(0.3\%)/\text{Ar}$ reducing atmosphere ($P_{\text{O}_2} = 10^{-8}$ atm at 1350°C) showed a piezoelectric constant (d_{33}) exceeding 500 pC/N. The piezoelectric properties of the Li_2CO_3 -added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ ceramics were comparable or superior to those of the same ceramic sintered in air. Also, grain growth was promoted even in Li_2CO_3 -added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ samples sintered at a lower temperature than 1350°C . The 3 mol% Li_2CO_3 -added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ ceramics fabricated at 1250°C in a reducing atmosphere exhibited the d_{33} approximately 580 pC/N.

MnCO_3 and Li_2CO_3 (3 mol%)-added $(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.95}\text{Sn}_{0.05})\text{O}_3$ ceramics were also studied not only to improve reduction resistance but also to control the stability of ferroelectric domain structure. In this case, Mn can act as an acceptor ion. The optimization of the amount of MnCO_3 addition was examined to increase the Q_m value. As a result, the samples sintered at 1250°C in a reducing atmosphere with 0.4 mol% of MnCO_3 addition showed the d_{33} value of 500 pC/N or higher. Furthermore, a relatively high mechanical quality factor Q_m value (approximately 600) was also achieved. These results suggest that Li, Mn co-doped $(\text{Ba,Ca})(\text{Ti,Sn})\text{O}_3$ ceramics sintered in a reducing atmosphere could be applied to high-power piezoelectric devices.

D-6:

Investigations of multi-scale regulation and in-situ characterization of high temperature piezoelectric ceramics

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High temperature piezoelectric ceramics with layered structures (including bismuth layer and perovskite layer), characterized by high Curie temperature (T_C : 650~1500 °C), are the key functional components of high temperature piezoelectric sensors, transducers and other devices at 500 °C and above, and are widely used in the fields of aerospace and nuclear energy for vibration monitoring and structural health management under high temperature, high radiation, complex vibration and other harsh environments.

Focusing on the bottleneck of low piezoelectric coefficient d_{33} of layered piezoelectric ceramics, the piezoelectric properties were improved by means of A/B co-doping and ion-pairing effect doping, and the structure-property relationship between structure and piezoelectric properties was systematically studied. d_{33} of bismuth layered $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ piezoelectric ceramic was increased from 7 pC/N to 40.2 pC/N.

In view of the lack of in situ high-temperature testing technology and equipment to study the service stability of high-temperature piezoelectric ceramics, based on the quasi-static and dynamic resonance methods, a multi parameter in-situ characterization technology and testing system from room temperature to 700 °C were developed, and the temperature stability of high-temperature piezoelectric ceramics was studied in depth, which promoted the practical application of piezoelectric materials and related devices in high-temperature fields.

Enhanced energy storage ceramic capacitors utilizing ferrorestorable polarization

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A self-powered system with a long lifetime offers an opportunity to develop a next-generation, standalone Internet of Things. Ceramic capacitors are promising candidates for energy storage components because of their stability and fast charge/discharge capability. Even for state-of-the-art capacitors, the energy density needs to be increased markedly. Improving breakdown electric fields provides a potential solution, but operations at such high fields relying on unchanged dielectric permittivity sacrifice the lifetime to some degree. Here, we report a ferrorestorable polarization engineering capable of enhancing effective permittivity over twice. Our experiments and *ab initio* calculations demonstrate that a defect dipole composed of 3d transition metal acceptors such as Cu^{3+} and oxygen vacancy in a prototypical ferroelectric BaTiO_3 ceramic is coupled with spontaneous polarization¹⁾. The resultant ferrorestorable polarization delivers an extraordinarily large effective relative permittivity beyond 7,000 with a high energy efficiency up to 89 %²⁾. Our work paves the way to realizing efficient ceramic capacitors for self-powered applications.

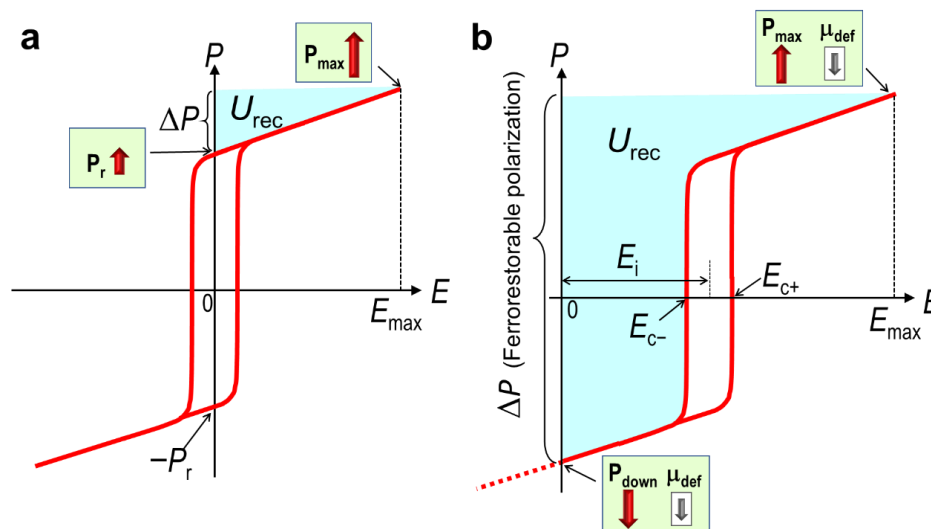


Fig. 1 | Ferrorestorable polarization²⁾. **a**, Typical P - E loop of ferroelectrics (pristine). **b**, Shifted P - E loop with an internal electric field (E_i) caused by the ground-state configuration of $\mu_{\text{def}} \parallel \mathbf{P}_s$ (controlled). The controlled sample has a large U_{rec} as a result of ΔP , which is termed ferrorestorable polarization. The interaction between μ_{def} and \mathbf{P}_s stabilizes the downwards polarization (P_{down}) at zero field, i.e., $P_0 = P_{\text{down}}$, because the P - E loop shifts to a positive field by the magnitude of E_i . E_i is defined as the average of E_{c+} and E_{c-} , that is, $E_i = (E_{c+} + E_{c-})/2$, where E_{c+} and E_{c-} are the electric fields at the extreme polarization switching currents in the positive and negative field sweeps, respectively.

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High performance BaTiO₃-based dielectric materials for capacitor applications

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BaTiO₃ (BT) is an important dielectric material, which have extensively been applied in electronic and electrical industries such as multilayer ceramic capacitors (MLCC), random access memory, sensors, and actuators. The harsh and extreme working environments have put forward strict requirements for temperature stability of dielectric materials, especially at higher temperatures. Intense efforts have been devoted to maintaining high dielectric constant over a wide range of temperature. It has been reported that forming solid solutions and constructing core-shell structures by doping with various additives are the most effective approaches to enhance dielectric temperature stability of BT based ceramics. As we all know, core-shell structure is a metastable state in thermodynamics, and will evolve to a more stable homogenous state depending on changes in producing conditions, such as sintering temperature, dwelling time, coating conditions, starting particle size, internal stress and thermal procedure.

In this work, the influence of different oxides, starting particle size and sintering conditions on the microstructures and dielectric properties of BaTiO₃-based ceramics were investigated to reveal the core-shell structure by high resolution transmission electron microscopy techniques with EDS analysis. The ion diffusion behavior plays a critical role in the formation and evolution of core-shell structure, and therefore influences the dielectric properties. When using more than 100 nm BaTiO₃ as starting powders and sintering for relatively shorter dwelling times (0.5 h-2.0 h), the chemical heterogeneity core-shell structure could be formed and retained owing to the limited diffusion behavior. However, when using 80 nm BaTiO₃ nanopowders and further extending dwelling times to 6 hours, the diffusion length was shortened and more driving energy was provided to prompt ion diffusion behavior, which made elements distribute uniformly and compositional inhomogeneity become homogenized. The double dielectric peaks in ϵ_r - T curves would become a single diffused peak with the collapse of core-shell structure.

Origin of High-Performance Energy Conversion in AgNbO₃ Based Ceramics: Structure Evidences

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Lead-free AgNbO₃ has attracted great attentions in ferroelectric physics due to its excellent energy storage and conversion properties. However, to develop lead-free energy conversion devices and the performance of AgNbO₃ based materials, the intrinsic mechanism of the high energy storage performance is still unclear, due to their complex lattice structure and doping engineering. In this report, a refined phase diagram was firstly constructed by studying the condensed-matter spectroscopies under the changes of chemical dopant, temperature and pressure fields. It is revealed that structural transformation is closely related to the antipolarity of the cation and the distortion of the oxygen octahedron, where the successive lattice transitions of AgNbO₃ are associated with displacive-type characterized by soft mode. Simultaneously, the synergistic study of phonon dynamics and electronic transition behaviors have been illustrated under the doping contents, the external temperature and stress fields. Furthermore, we confirmed that the main factors determining the excellent energy conversion performance of AgNbO₃ ceramics are including the large remanent polarization for enhancing the energy storage density, the wide bandgap and small crystalline grain for improving the intrinsic breakdown strength, as well as the coexistence of polar and non-polar phase for reducing FE-AFE energy barrier. Our work provides a full strategy to explore the physical mechanism of the polar-nonpolar structural transition and the intrinsic correlation between the lattice structure and the functionalities, which would promote the development of lead-free energy storage ceramic.

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Growth of Metal-free Perovskite-type Ferroelectric Crystals and Their Ferroelectric Properties

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Recently, it was reported that metal-free perovskite-type crystals such as (MDABCO)(NH₄)I₃ crystals were true ferroelectric crystals with larger spontaneous polarization (P_s) of almost 20 μC/cm², very similar to P_s of BaTiO₃ (P_s ~ 26 μC/cm²)¹. Until today, various metal-free ferroelectric crystals were reported by many researchers, but most of crystals showed very low spontaneous polarization below 5 μC/cm². So, we believe that it is very important to confirm P_s value (almost 20 μC/cm²) of (MDABCO)(NH₄)I₃ crystals. In this study, two oriented (MDABCO)(NH₄)I₃ crystals along to <111> and <100> directions were grown by conventional crystal growth method at room temperature. Then, crystal structure of these crystals was measured. At the same time, various ferroelectric related properties were measured. These detailed results will be reported at the conference.

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Effect of Reaction Accelerator on Single Crystal Barium Titanate Fine Particle Synthesis

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1. Introduction

Barium titanate (BT), one of the important electro-ceramic materials, is used to multi-layer ceramic capacitors (MLCCs), electro-optical (EO) devices, sensor devices and so on. In particular, BT fine particles are expected to be applied to miniaturization of ceramic capacitors and as ordered accumulations. Considering that the space is filled with only one type of solid, its shape is limited to cubic, rhombic dodecahedrons, and so on. The case of liquid phase crystal growth, the particle shape can be controlled by adsorbing the additive on specific surface of the crystal. Therefore, in this study, we describe the results of examining the effect of synthetic conditions on the shape of BT fine particles.

2. Experimental Procedure

BT fine particles were fabricated by hydrothermal synthesis method, in this study. The precursor solution is mixture of barium hydroxide, titanium lactic acid complex, sodium hydroxide, activator and surfactant. The Ba/Ti ratios was controlled exactly 1.0. During the heating of solution, it was mechanical stirring. After the synthesis, the samples were washed and classification. Particle synthesis and particle shape evaluation were performed as in previous reports.¹

3. Results and Discussions

In this study, tert-butylamine (tBA) and tert-amylamine (tAA) was used as a reaction accelerator. We can see from the figures, microparticles grow independently of the type of reaction accelerator. In addition, although not shown here, it was confirmed from the results of XRD observation that the fine particles synthesized by tBA were perovskite-type BT, and high-resolution image analysis indicates that the microparticles are single crystals surrounded by {100} planes and belong to the tetragonal system with a lattice constant of approximately 0.4 nm. The oxygen octahedral structure, which is the basic skeleton of the perovskite structure, is formed by dissociating lactic acid from the titanium complex. Considering the molecular structure and Pauling's electronegativity, tBA, which is smaller and has higher electrical symmetry, is expected to have a greater reaction-promoting effect. In fact, the synthetic particle size of tBA is larger than that of tAA. Therefore, we think that tBA is suitable as a reaction accelerator. However, there is a problem that the particle shape is not uniform.

Acknowledgements

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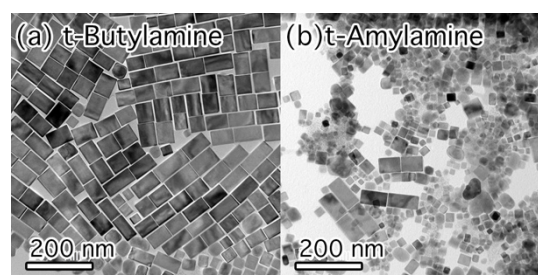


Fig. Shape changes of synthesis BT fine particles by a reaction accelerator.

Ferroelectric and piezoelectric properties in simple binary compounds

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First-principles calculations are performed to investigate ferroelectricity in some binary compounds, including bilayer two-dimensional (2D) hexagonal boron nitride (h-BN) and rocksalt oxide family (AO with A=Cd, Ba, Sr, Ca, Mg, etc.) via the application of biaxial strain. The origins of such unconventional ferroelectricity are discussed from the lattice perspective and electronic structure properties. These findings have the potential to motivate the study of simple materials possessing ferroelectricity and piezoelectricity that may lead to novel devices.

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Huge Piezoelectric Response of LaN-based Superlattices.

Minglang Hu, et al., and W. Ren, *ACS Appl. Mater. Interfaces* 12, 49805 (2020)

Angle-resolved polarized Raman mapping on a compositionally graded $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$

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Explaining the “properties and functions of materials” in terms of “atomic arrangement and various inhomogeneous structures” is a fundamental challenge for developing ferroelectric oxides. Dielectricity, one of the most fundamental properties of matter, can be explained by long-wavelength polar lattice vibrations and dipole relaxations, accompanied by the bias of electricity. Therefore spectroscopic methods such as Raman spectroscopy are used to investigate the origin of the property. Using a recently advanced angle-resolved polarized Raman mapping, we investigate the correlation between the phase boundary and the giant dielectric responses in relaxor $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ and ferroelectric PbTiO_3 solid solution (PMN-*x*PT) system using a compositionally graded crystal [1].

Angle-resolved polarization Raman mapping, X - θ mapping, which was developed by incorporating information of polarization of light θ into Raman mapping, was performed [2]. A schematic diagram of the experiment is shown in FIG. 1(a), and the X - θ contour map on the peak at 750 cm^{-1} is shown in FIG. 1(b). Gradual changes in polarization direction are visualized by Raman mapping the crystal. And by analyzing quasi-elastic scattering from dipole relaxation, the origin of the enormous dielectric response found in PMN-*x*PT can be understood as follows: The mesoscopic ferroelectric domains, which are composed of polar nano regions, become smaller and slowly fluctuate near the phase boundary, resulting in an enormous dielectric response, which is the characteristic of relaxor-ferroelectric solid solution system and the difference from other ferroelectrics.

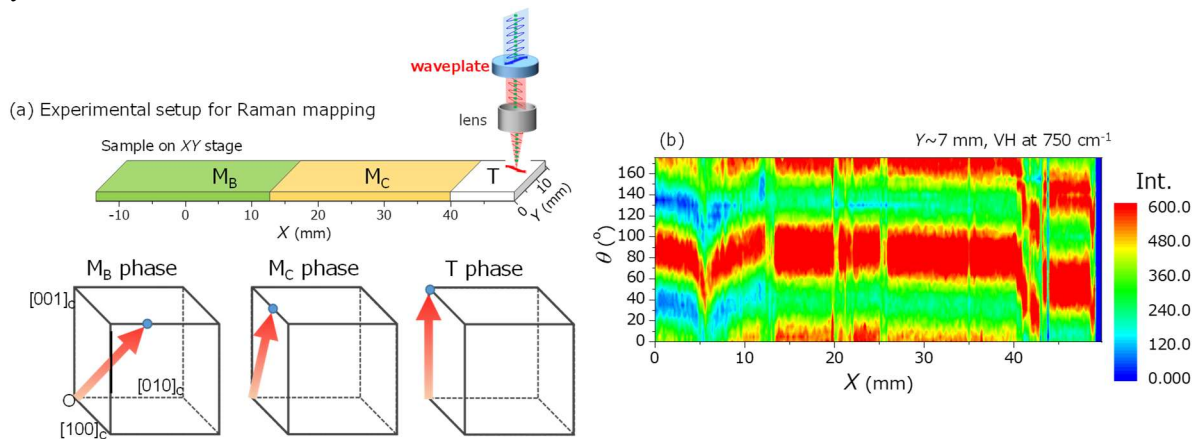


FIG. 1 (a) Schematics of sample setting in Raman microscope with the relationship between structure and polarization direction. M_B , M_C , and T denote monoclinic (type B), monoclinic (type C), and tetragonal crystal structures, respectively. (b) An X - θ Raman mapping image colored by the area around 750 cm^{-1} of the spectra measured under the conditions $Y \sim 7\text{ mm}$.

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Flexible Lead-free Piezoelectric Arrays for High-Efficiency Wireless Ultrasonic Energy Transfer and Communication

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Implantable medical electronics (IMEs) are now becoming increasingly prevalent for diagnostic and therapeutic purposes. Despite extensive efforts, a primary challenge for IMEs is reliable wireless power and communication to provide well-controlled, therapeutically relevant effects. Ultrasonic energy transfer and communication (UETC) employing traveling ultrasound waves to transmit energy has emerged as a promising wireless strategy for IMEs. Nevertheless, conventional UETC systems are rigid, bulky, and based on toxic lead-based piezoelectric materials, raising efficiency and safety concerns. Here, we present a novel transcutaneous UETC system based on a two-dimensional flexible lead-free piezoelectric array (f-LFPA) that hybridizes high-performance (piezoelectric coefficient $d_{33} \approx 503 \text{ pC N}^{-1}$) (K,Na)NbO₃-based eco-friendly piezo-units with soft structural components. The newly developed lead-free piezo-unit exhibits submicron ferroelectric domains and superior energy harvesting figures of merit ($d_{33}g_{33} \approx 20000 \times 10^{-15} \text{ m}^2 \text{ N}^{-1}$), resulting in the prepared f-LFPA demonstrating a high output voltage of 22.4 V, a power density of 0.145 W cm^{-2} , and a signal-to-noise ratio of more than 30 dB within the FDA safety limits, while maintaining the flexibility for wide-angle receiving. Further ex vivo experiment demonstrates the adequate power supply capabilities of the f-LFPA and its possible application in future implantable eco-friendly bioelectronics for diagnostics, therapy, and real-time monitoring.

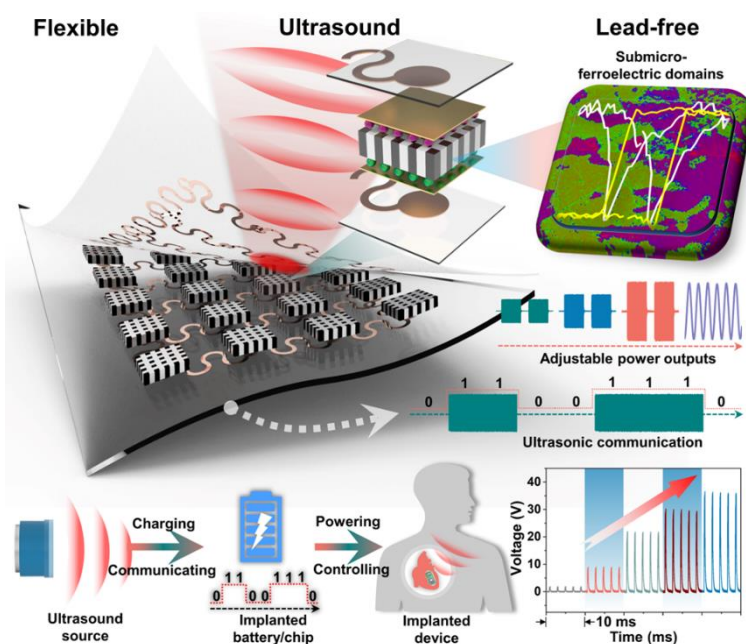


Fig. 1 Schematics and design of the flexible lead-free ultrasonic device.

RTIC phenomenon on AD process and its application to dielectric / ferroelectric thick films

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Aerosol Deposition (AD) method[1]-[4] is a unique approach for metal and ceramic coating, where solid state submicron metal and ceramics particles are accelerated by gas flow up to 100 - 500 m/s and then impacted onto a substrate. AIST found interesting consolidation phenomenon of ceramic particles in this method over 20 years ago. During collision of fine particles and interaction with substrate, these ceramic particles, not only for oxide materials but also for non-oxide materials, formed thick, dense and hard ceramic layers at room temperature. No additional heating for solidification of ceramic powder was required. We named this phenomenon “Room Temperature Impact Consolidation (R.T.I.C)” [2],[4]. Consolidated ceramic powder with R.T.I.C. via AD method can be called as a high density binder less ceramic green. It can form rapidly a thick dense, and uniform ceramic layer at room temperature without additional heating for solidifying starting powders, even in low vacuums, using relatively cheap and simple production facilities. Metal-based optical scanning devices, tube-type ultrasonic motors, piezoelectric energy harvesters and energy storage using dielectric / ferroelectric AD thick films have been developed for sensor & actuator and energy device applications.[3]

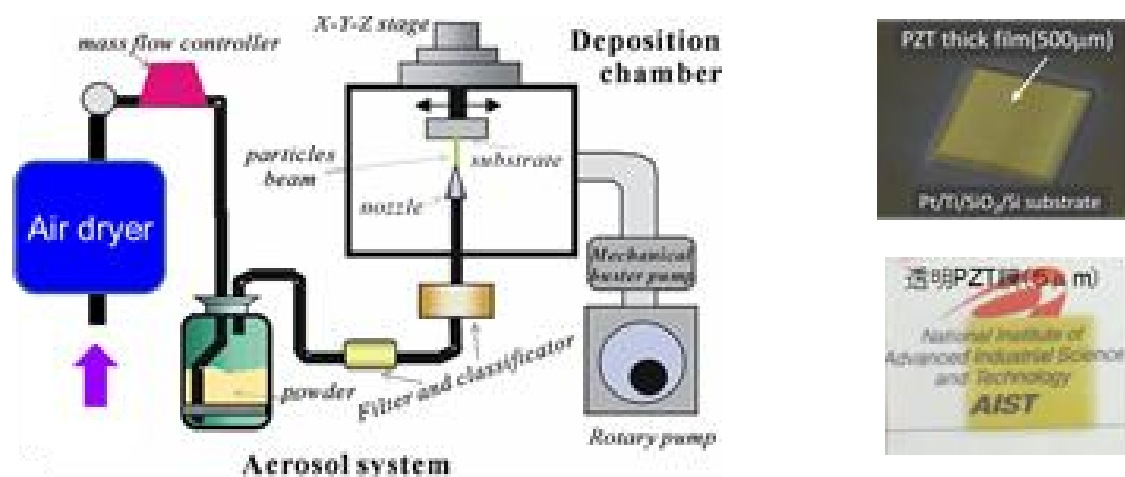


Figure 1 : The configuration of AD machine and the deposition example of AD method (PZT thick film on glass and Si wafer)

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Ferroelectric gate thin films transistors with Y-doped Hf-Zr-O gate insulator and In-Sn-O channel

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HfO₂-based ferroelectric materials, whose ferroelectricity was reported in 2011[1], have attracted much attention as candidates for future electronics applications, such as nonvolatile memories and steep-slope MOSFETs. One of the most promising applications is a ferroelectric-gate field effect transistor, called FeFET or FGT. In this device, a ferroelectric material is used as a gate insulator, which results in hysteresis characteristics in the transfer curve. In addition, because of the spontaneous polarization, the ferroelectric gate insulator can induce large charge density so that conductive oxide such as indium-tin oxide (ITO) can be used as a channel of the FGT [2]. One of the advantages of ITO channel FGT is its low “on”-resistance and low resistivity of local metallization, especially when it is used in NAND strings. ITO channel FGT is also applicable for 3D NAND structures.

In this talk, we report ferroelectric properties of Y-doped Hf-Zr-O (Y-HZO) thin films prepared by chemical solution deposition (CSD). Figure 1 shows polarization – electric field (P-E) hysteresis loops of Y-HZO films prepared by CSD with an Y concentration of 3.2 % [3]. The film was annealed at 800 °C for 3 min at a reduced pressure of 50 Pa. Clear ferroelectric properties were observed with a remanent polarization, P_r , of approximately 12 $\mu\text{C}/\text{cm}^2$. The P_r values estimated from P-E measurements are plotted in Fig.2 as a function of Y concentration and it was found that the maximum P_r was obtained when the Y concentration is around 3 %. Next, we apply the CSD ferroelectric Y-HZO film to FGT. To fabricated FGTs, ITO layer with a Sn concentration of 5% is fabricated by CSD on the Y-HZO gate insulator. Figure 3 shows transfer curve obtained for ITO/Y-HZO structure FGT. A clear hysteresis with a memory window of as large as 10 V was observed. The details will be presented at the conference.

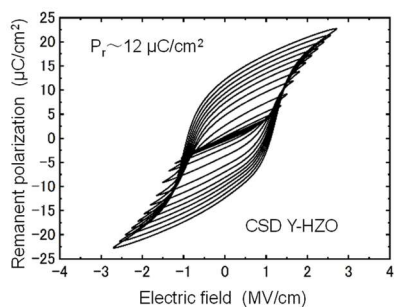


Fig.1 P-E loops of Y-HZO films Prepared by CSD

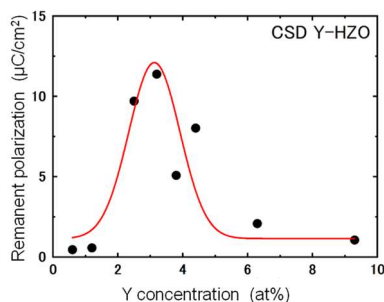


Fig.2 Remanent polarization as a function of Y concentration

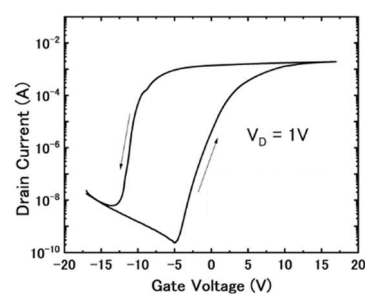


Fig.3 Transfer curve of ITO/Y-HZO FGT.

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F-4:

Fabrication and characterization of La-doped HfO₂ ferroelectric films on SrTiO₃ single crystal substrates.

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Several doped-hafnium oxide films have recently been discovered to have ferroelectric properties. These films are peculiar because unlike conventional ferroelectric materials, their ferroelectricity does not diminish with decreasing film-thickness. In the present work, La-doped hafnia films were deposited by pulsed laser deposition on La_{2/3}Sr_{1/3}MnO₃ buffered SrTiO₃ (100) substrates. The crystal structure and ferroelectric properties were studied with varying deposition and annealing conditions. Films deposited at high-temperature showed good crystallinity but also exhibited more leakage current. On the other hand, films deposited at room-temperature which were later annealed in a nitrogen atmosphere showed low leakage current, but had poorer crystallinity. Various aspects of optimization of the fabrication process, and the parameters critical for enhanced ferroelectric properties are discussed.

Energy-storage performance of antiferroelectric Pb(Zr,Hf)O₃ films fabricated by sol-gel spin coating

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Antiferroelectric (AFE) materials, such as PbZrO₃ (PZ) and AgNbO₃, are competitive candidates for the energy-storage application, owing to their large saturated polarization and small remanent polarization^[1]. PbHfO₃ (PH), with a similar crystal structure of PZO, has also attracted great attention in recent years and comparable recoverable energy (W_{re}) with PZO has been reported^[2-3]. However, investigation on the microstructure and energy-storage performance of Pb(Zr_{1-x}Hf_x)O₃ (PZH) is quite limited. In this study, PZ, PH and PZH films with the Zr/Hf ratio of 90/10, 80/20 and 50/50 were fabricated on Si substrates using the sol-gel spin coating method. The P - E hysteresis loops showed that all the samples exhibited similar remanent polarization and low leakage current. The maximum polarization increased with Zr content, indicating the enhancement of W_{re} in Zr-rich films. The energy efficiency was more affected by the existence of Hf than the amount of Hf. As a result, the PZH (90/10) film with the highest W_{re} of 11.3 J/cm and a larger energy efficiency of 55%, measured at 800 kV/cm, was obtained in this study. This suggests that the Hf-doped PZ film is promising for the energy-storage application.

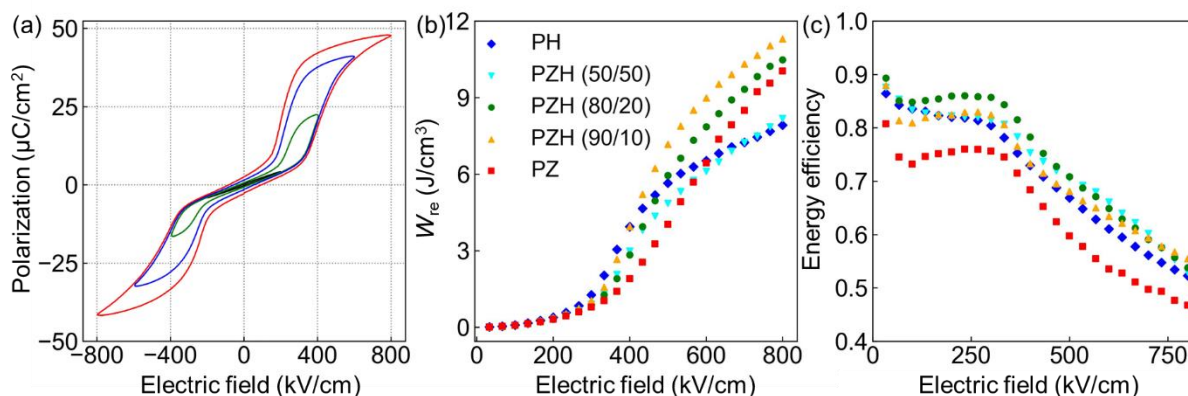


Fig. 1 (a) The P - E hysteresis loop of PZH (90/10), measured at 10kHz. (b) Recoverable energy density (W_{re}) and (c) energy efficiency of the fabricated films, changed with the applied electric field.

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Low temperature deposition of BiFeO₃ films on base metal foils for piezoelectric applications

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Piezoelectric BiFeO₃ (BFO) films were successfully prepared on 304 stainless steel (SS) and Ti foils via RF-magnetron sputtering at 450 °C. By adopting a LaNiO₃/Pt/Ti tri-layer bottom electrode, a predominant (100)-orientation and a dense fine-grained morphology were achieved in these BFO films, resulting in good ferroelectric and dielectric properties targeted for piezoelectric applications. These properties include a large remnant polarization ($P_r \sim 68 \mu\text{C}/\text{cm}^2$ for BFO-SS, $P_r \sim 72 \mu\text{C}/\text{cm}^2$ for BFO-Ti), a sizable self-bias ($E_{br} \sim 158 \text{ kV}/\text{cm}$ for BFO-SS, $E_{br} \sim 145 \text{ kV}/\text{cm}$ for BFO-Ti), and very decent dielectric properties ($\epsilon_r \leq 270$, $\tan \delta \leq 0.026$ in 1 kHz-1MHz). Large transverse piezoelectric coefficients of $|e_{31,j}| \sim 2.2 \pm 0.2 \text{ C}/\text{m}^2$ and $|e_{31,j}| \sim 1.7 \pm 0.1 \text{ C}/\text{m}^2$ were achieved in patterned BFO-Ti and BFO-SS cantilevers, respectively. A decent direct transverse piezoelectric coefficient ($\sim 1.07 \pm 0.03 \text{ C}/\text{m}^2$) was also achieved in BFO-SS based prototype piezoelectric energy harvesters. These results suggest that BiFeO₃ films have a great potential for applications in lead-free, metal-based piezoelectric micro-electro-mechanical systems (Piezo-MEMS).

Extraordinarily large domain switching contribution in epitaxial (K, Na)NbO₃ films

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(K, Na)NbO₃ (KNN) as a promising alternative to the state-of-art PZT has drawn enormous attention due to high curie temperature and excellent piezoelectric response.^[1] The piezoelectric response is derived from intrinsic lattice deformation and extrinsic domain wall motion and domain volume variation. Despite significant efforts on the piezoelectric response in KNN films, the ratio of intrinsic and extrinsic contributions remains elusive. Yet, *in-situ* time-resolved synchrotron X-ray diffraction can offer assistance to clarify the origin of large piezo-response.^[2, 3] Here, we successfully grew the KNN films in monoclinic phase, of which sole (001) domain in KNN (001)_{pc} films, but ($\bar{1}11$) and (111) domains in KNN (111)_{pc} films. Notably, KNN (111)_{pc} films manifest the fast and reversible domain switching, whose contribution to the piezoelectric response surpasses 100% under the moderate bias.

The piezo-response is intimately relevant to the orientation of the domains. Fig. 1 exhibits the field-induced strain for KNN (111)_{pc} films. The intrinsic and extrinsic piezoelectric responses are highly relying on the applied field; the moderate bias enhances the extrinsic response via domain switching, whereas the intrinsic response emerges only over it. The extrinsic contribution to $d_{33,f}$ exceeds 100% due to the slightly negative intrinsic response under it. Fig. 2 shows the time evolution for the (111)_{pc} KNN film under electric field of 36 kV/cm. The domain switching in the (111)_{pc} film is fully reversible. The domain switching is saturated during the interval of the voltage pulse of 50 μ s. Both time of inducing the domain switching and relax back to the original state after releasing the bias was around 30 μ s.

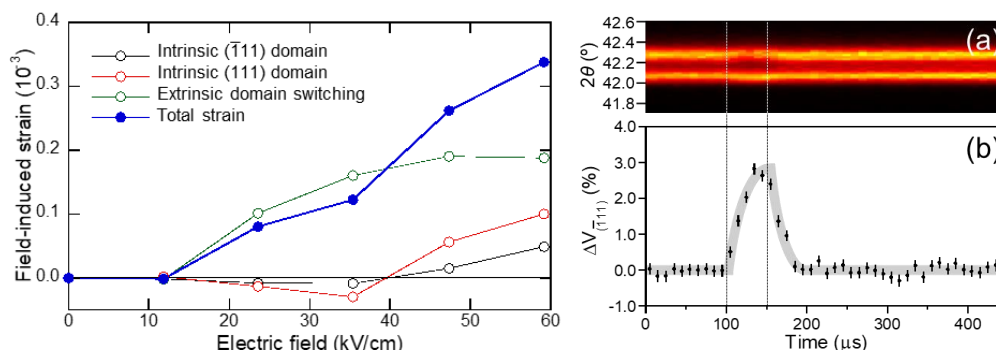


Fig. 1 Field-induced strain for KNN (111)_{pc} films. Fig. 2 Time evolution of domain switching for KNN (111)_{pc} film under 36 kV/cm.

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Intrinsic properties of domain walls of BiFeO₃

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Embedded in the bulk of an insulating material, ferroelectric domain walls have the capability of being (re)created, displaced, erased, and altered in their spatial configurations and electronic characteristics. In recent years, domain walls themselves have attracted extensive attention, due to rich intriguing physics and novel ideas of designing nanoelectronics based on these interfaces. Intrinsic properties of domain walls, e.g., domain wall conductivity, domain wall type, and domain wall width, are thus particularly important in understanding characteristics of ferroelectrics involving domain walls. In recent years, we obtained domain wall conductivity ($> 10 \mu\text{A}$) more than one order of magnitude higher than previously reported maximum value ($\sim 0.6 \mu\text{A}$). However, reliable routine to design a giant conductivity for random domain walls is lacking. Such discussion is in great demands. Moreover, these free-carrier compensated non-180° CDW may show a thickness as thin as ~ 2 unit cells, with a Néel-like character, unlike prior beliefs.

Domain walls are interesting themselves, containing rich physics and are related to many new phenomena, as well are critical tangling with their macroscopic performances. Interacting with the insulating bulk within the piezoelectric materials, conductive domain walls would induce peculiar bulk property response of these materials, for example decoupling of lattice strain and domain wall motion induced strain in both the frequency and field amplitude domains. These are surprising and unexpected with fundamental implications for a broad range of materials as well as for device design. The intrinsic properties of domain walls worth more thorough investigations.

Phase Transformation, Interfacial Coupling and Giant Charge-discharge Efficiency of BaTiO₃@PVP/PVDF Nanocomposite Films

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Abstract: The introduction of BaTiO₃ (BT) nanoparticle modified on the surface of Polyvinylpyrrolidone (PVP) into semi-crystalline polyvinylidene fluoride (PVDF) matrix can effectively enhance the interfacial coupling, charge-discharge efficiency and energy storage performance of inorganic-organic nanocomposite films. In this study, compared with the BT/PVDF composite films, the interfacial coupling ability of BT@PVP/PVDF has been greatly improved, which is attributed to the bridge effect of PVP. Besides, when the BT@PVP is filled with 5 vol%, the energy density, the breakdown field strength, and the efficiency are respectively increased by 257.8% (18.39 J/cm³ vs 5.14 J/cm³), and 51.5% (458.3 MV/m vs 302.5 MV/m) and 47.56% (79.8% vs 54.08) compared with BT/PVDF composite films. Outstandingly, the remarkable charge-discharge efficiency is the higher value than reported so far. Furthermore, the enhancements of these performance are importantly related not only to the interfacial coupling of inorganic-organic nanocomposite films, but also to the increase of the $\alpha+\gamma$ phases (the $\alpha+\gamma$ phases content of 5 vol.% BT@PVP/PVDF increased by 7.82% compared with BT/PVDF composite films). This work demonstrates a possible mechanism for the improvements of the PVDF-based surface-modified inorganic ceramic nanocomposites' energy storage performance.

Keywords: interface couple, inorganic-organic, BT@PVP/PVDF composite film

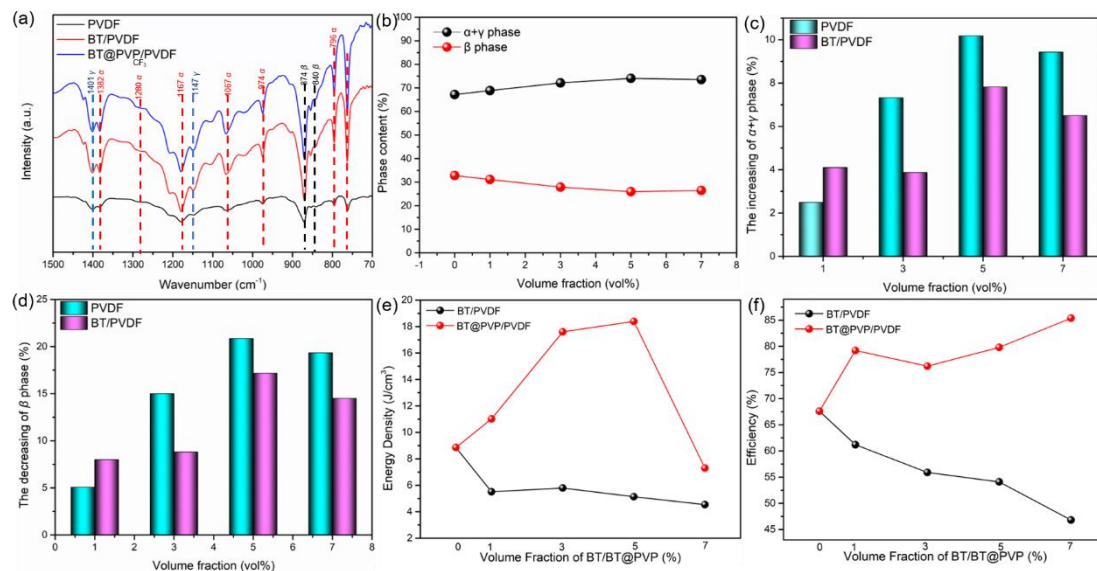


Fig. 1 (a) The Fourier transform infrared spectra of pure PVDF films, 5 vol.% BT/PVDF and 5 vol.% BT@PVP/PVDF nanocomposite films. (b) Phase content analysis of BT@PVP/PVDF composite films for $\alpha+\gamma$ phases and β phase. (c) $\alpha+\gamma$ phases increasing of and (d) β phase decreasing of BT@PVP/PVDF nanocomposite films comparing with pure PVDF films and BT/PVDF nanocomposite films. (e) Energy density and (f) Discharge-charge efficiency of BT/PVDF and BT@PVP/PVDF composite films.

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Crystal structure and microwave dielectric properties of $\text{Ba}_4(\text{Sm}_{1-x}\text{Ce}_x)_{9.33}\text{Ti}_{18}\text{O}_{54}$ solid solutions

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The rapid development of the fifth generation (5G) wireless communication technology has made greater performance demands for microwave components such as dielectric resonator and dielectric filter. Microwave dielectric ceramics, as the core materials of microwave devices, have been a research hotspot. In order to meet the requirements of microwave devices, superior performance of microwave dielectric ceramics must require, have medium/high relative dielectric constants (ϵ_r) to realize the integration and miniaturization of microwave devices, have high quality factor ($Q \times f$) to enhance the frequency selection characteristics of the device, have near-zero temperature coefficient of resonance frequency (τ_f) to obtain high temperature stability. The $\text{Ba}_4\text{Sm}_{9.33}\text{Ti}_{18}\text{O}_{54}$ ceramic system belongs to the tungsten bronze structure, which is composed of titanium-oxygen octahedra interconnected by means of shared vertices, forming three kinds of voids in space: the quadrilateral position A1 occupied by Sm^{3+} , the pentagonal position A2 occupied by Ba^{2+} with a slightly larger ionic radius, and the triangular void not occupied by ions. The uniformly ordered arrangement of Sm^{3+} and Ba^{2+} in the voids A1 and A2 arrangement reduces the internal stresses making the $Q \times f$ value maximum. In order to further improve the $Q \times f$ value, many doping modification studies were carried out.

In this work, $\text{Ba}_4(\text{Sm}_{1-x}\text{Ce}_x)_{9.33}\text{Ti}_{18}\text{O}_{54}$ ($x = 0.000, 0.125, 0.250, 0.375, 0.500$) microwave dielectric ceramics were prepared by the conventional solid-phase method. The effects of Ce^{3+} substitution for A-site Sm^{3+} on the microscopic morphology, crystal structure and microwave dielectric properties of the ceramics were investigated. A single tungsten-bronze phase was obtained for all the studied components. With the increase of Ce^{3+} substitution, the cell volume gradually increases, the internal stress inside the crystal decreases, the stability of the crystal structure increases, the value of quality factor $Q \times f$ increases, the relative permittivity ϵ_r increases, and the temperature coefficient τ_f of resonant frequency shifts toward positive values. At $x = 0.250$, $\text{Ba}_4(\text{Sm}_{1-x}\text{Ce}_x)_{9.33}\text{Ti}_{18}\text{O}_{54}$ ceramics exhibit excellent microwave dielectric properties when sintered at 1325 °C for 4 h: $\epsilon_r = 81.04$ (4.914957 GHz), $Q \times f = 9013$ GHz, and $\tau_f = -0.61$ ppm/°C.

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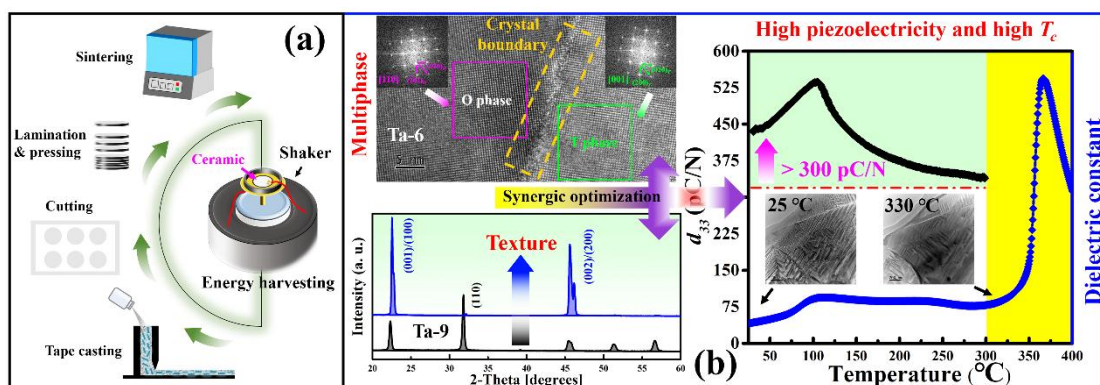
Ultrahigh energy harvesting properties in temperature insensitive eco-friendly high performance KNN-based textured ceramics

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High-performance potassium sodium niobate (KNN) based texture ceramics and its potential application on energy harvesting device are the first time to be presented in this work. A series of lead-free $0.99\text{K}_{0.5}\text{Na}_{0.5}\text{Nb}_{(1-x)}\text{Ta}_x\text{O}_3-0.01\text{Bi}(\text{Ni}_{2/3}\text{Nb}_{1/3})\text{O}_3$ [KNNTa-BNN] piezoceramics are designed and prepared to search for good comprehensive properties. The comprehensive performance of KNN-based piezoelectric ceramics have reached a new level in T-Ta-9 ($d_{33} \sim 435$ pC/N, $k_p \sim 71\%$, $T_c \sim 360^\circ\text{C}$) via the synergy of textured structure ($f_{(001)} > 94\%$) and multiphase coexistence (O-T) near room temperature. Based on the advanced *in-situ* TEM and PFM analysis, it is found that the regular large-scale domains and corresponding poling patterns of the T-Ta-9 can be well maintained at relatively high temperatures (180-330 $^\circ\text{C}$), resulting in excellent temperature stability. Hence, the d_{33} value of T-Ta-9 can be maintained above 300 pC/N over a wide range from room temperature to 300 $^\circ\text{C}$, which rarely happens in the previously reported KNN-based materials. More importantly, the piezoelectric circular diaphragm (PCD) vibration energy harvester based on the T-Ta-9 texture ceramic possesses high output voltage (~ 13 V) and output power (~ 3 mW), and can still maintain above 60% after heated at 200 $^\circ\text{C}$ for 30 min. This work represents a significant advancement in lead-free piezoelectric energy-harvesting field (especially for high temperature applications) and can provide guidelines for future efforts in this direction.



Characteristics of Chemical Bonding and Thermal Behavior of Atoms in Prototype Structure of Double Perovskite-type Antiferroelectric Oxides

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The solid solution $\text{Pb}(\text{Yb}_{1/2}\text{Nb}_{1/2})\text{O}_3$ (PYN) is a double perovskite-type oxide with an ordered arrangement of Yb and Nb atoms at the *B*-site. PYN undergoes the antiferroelectric phase transition at 575K, and exhibits an orthorhombic structure with the space group of *Pbnm* at room temperature. The phase transition should not be discussed only by the tolerance factor t ($t = 0.957 < 1$), but the intrinsic origin should be discussed from the lattice instability in the prototype structure.

In this study, we have revealed the structural characteristics on the lattice instability in the paraelectric phase of PYN, which causes the antiferroelectric phase transition, by visualizing the electron charge density distribution from the synchrotron radiation X-ray powder diffraction (SXRD) data. The Pb atom was off-centered at 12 equivalent sites at random in the $\langle 110 \rangle$ directions in the paraelectric phase, while the Pb atom was ordered at one of the off-centered sites in the antiferroelectric phase as shown in Fig 1. The electron charge density analysis of the paraelectric phase revealed the hybridization of atomic orbitals between the Nb and surrounding O atoms, while the Yb atom was fairly ionic. The thermal motion of the O atom was significantly anisotropic. The electron charge density distributions around the O atoms were extended toward the directions perpendicular to the Yb-O bond. We consider that the lattice instability to the antiferroelectric phase is due to the ionic Yb ion, which causes the tilting of the oxygen octahedra at the phase transition. The ordered direction of the off-centered Pb atom should be associated with the direction of the local polarization in the antiferroelectric phase.

The detail of crystal structures and the origin of the antiferroelectric phase transition will be discussed in presentation.

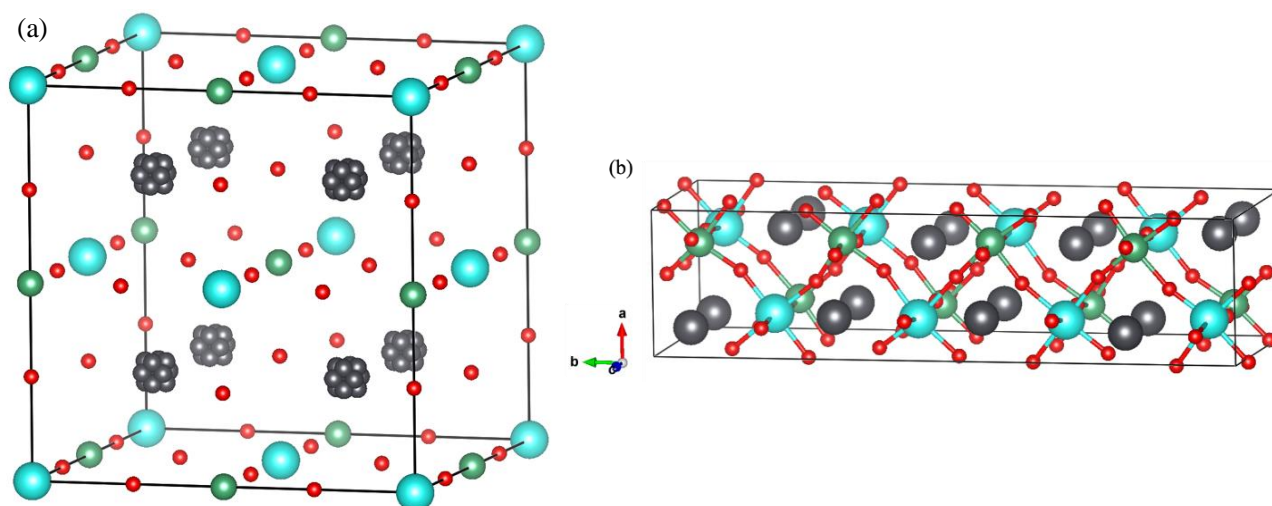


Fig. 1 Crystal structures of PYN obtained by the Rietveld analysis; (a) paraelectric phase at 673 K, and (b) antiferroelectric phase at room temperature.

Tunable vertical ferroelectricity and domain walls by interlayer sliding in β -ZrI₂

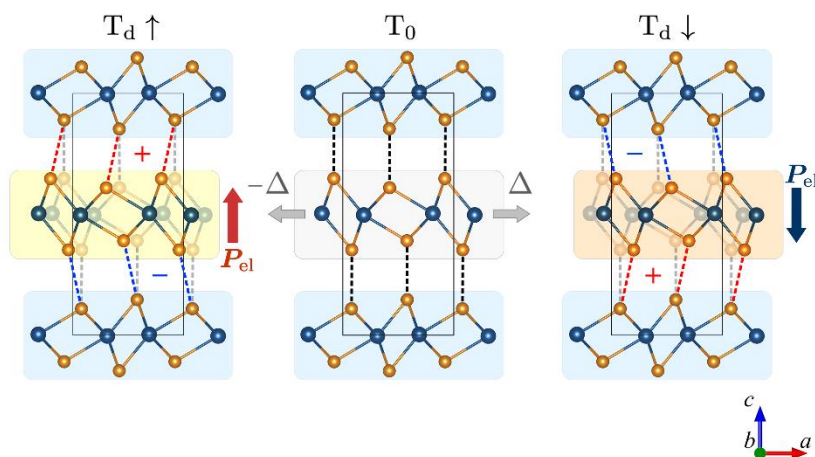
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Layered transition-metal dichalcogenides have gained enormous attention because of their ferroelectric properties caused by in-plane ionic displacements. In this study, we predict robust vertical ferroelectricity upon interlayer sliding in beta-ZrI₂, a sister material of polar semimetals MoTe₂ and WTe₂. Based on extensive first-principles calculations, we elaborate the microscopic origin of electric polarization in ZrI₂ and investigate the variety of ferroelectric domain boundaries, that alongside with the semiconducting behavior and a small switching barrier can hold promise for novel ferroelectric applications.



Crystal Structure of $\text{KNbO}_3/\text{BaTiO}_3$ Core-Shell Nanocomposite Particles Studied by Synchrotron Radiation X-ray Diffraction

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Considering environmental issues, the development of high-performance lead-free ferroelectrics to replace lead-based ferroelectrics is an urgent issue. Lead-free ferroelectric with an artificial superlattice have a remarkable effect on improving the dielectric properties of lead-free. For example, heteroepitaxial thin films exhibit excellent dielectric properties. A lattice-strain-layer appears at the interface due to the lattice mismatch, which enhances the polarization. Our group has attempted to form a lattice-strain-layer in a nanocomposite ceramic particle with a core-shell structure. We have revealed that the lattice-strain-layer governs the dielectric property of the nanocomposite particle.

In this study, we demonstrated the temperature variation of the crystal structure of lattice-strain-layer in the $\text{KNbO}_3(\text{KN})/\text{BaTiO}_3(\text{BT})$ nanocomposite particles. The nanoparticles were prepared by the solvothermal method. The core of the particle is BT with a diameter of 300 nm, and the shell is KN heteroepitaxially covering the BT core. The synchrotron radiation X-ray powder diffraction revealed the crystal structure of the nanoparticle. The Rietveld method was used for the structural refinement. However, the powder pattern of the KN/BT nanoparticles at 800 K could not be refined with a combination of single structural components with a cubic structure of KN and BT, respectively. Two cubic components models with different lattice parameters for KN and BT respectively gave the best fitting result in the Rietveld refinement. The schematic diagram of the one KN/BT nanoparticle is shown in Fig. 1, where the four components in all are arranged. It can be confirmed that the lattice-strain-layer exists at the interface in the combination of cubic KN and cubic BT. The crystal symmetry of KN at 600 K is tetragonal, while BT is cubic. In the case the crystal system is different, the lattice-strain-layer at 600 K is revealed to exist in a wider area near the interface.

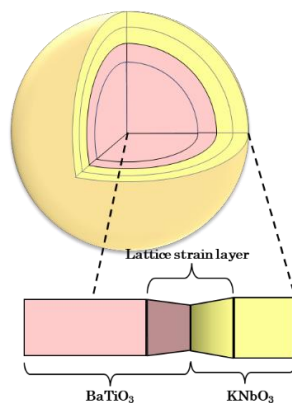


Fig. 1 Schematic of heteroepitaxial KN/BT nanocomposite particle with core-shell structure.

Brillouin Scattering Study of Ferroelastic Phase Transition of Lead Phosphate Single Crystal

Adil Emin^{1,2,*}, Anwar Hushur², and Seiji Kojima³

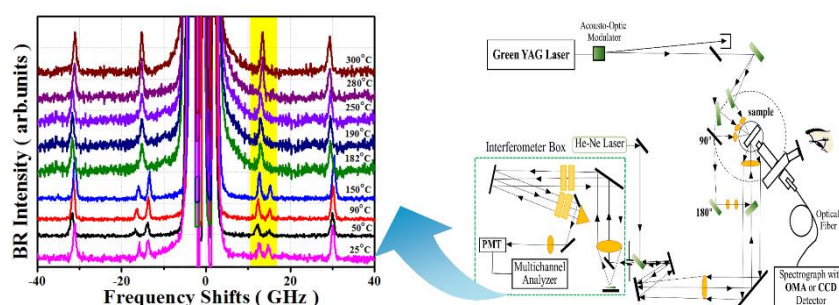
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Graphical abstract



Abstract

Brillouin scattering spectroscopy plays a vital role in the study of various kinds of phase transitions and critical phenomena in condensed matters. For structural phase transitions in crystalline solids, Brillouin scattering experiments provide detailed information on elastic and gigahertz dynamical properties. We have used the Brillouin scattering technique to investigate the pure ferroelastic phase transition of a lead phosphate $\text{Pb}_3(\text{PO}_4)_2$ crystal at high temperatures. The ferroelastic domain configuration confirms previous identifications of the symmetry elements lost at the transition. The temperature dependence of acoustic mode frequency indicates that these lead phosphate $\text{Pb}_3(\text{PO}_4)_2$ undergoes a first-order ferroelastic phase transition at $T_C = 182$ °C from the paraelastic high temperature (HT) trigonal (rhombohedral) phase (the space group $R\bar{3}m-D_{3d}^5$) to the ferroelastic low temperature (LT) monoclinic phase (the space group $C2/c-C_{2h}^6$) caused by L-point instability in the trigonal phase, which doubles the volume of the unit cell. Neither soft acoustic mode and central peak was observed in the vicinity of T_C . The results show that the staggered ferroelastic phase transition is order-disorder type.

Keywords: $\text{Pb}_3(\text{PO}_4)_2$, ferroelastic phase transition, Brillouin scattering, elastic anomaly, high temperature.

Structural evolution and electrical properties of BFO-BT based high temperature lead-free piezoelectric ceramics

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Bismuth ferrite-barium titanate (BiFeO₃-BaTiO₃, BFO-BT)-based piezoelectric ceramic is one of the most important lead-free ferroelectric material systems, because it has high Curie temperature, good temperature stability and low cost of raw materials. On the other hand, the relative small piezoelectricity limits the practical application of the BFO-BT based ceramics. The morphotropic phase boundary (MPB), a ferroelectric phase transition boundary, is considered as the origin of high piezoelectricity of the Pb(Zr,Ti)O₃ (PZT) piezoelectric materials. Based on the MPB effects in the classical PZT systems, tailoring a MPB by chemical modification becomes a promising strategy to design high-performance BFO-BT based lead-free piezoelectric ceramics.

In this work, the (1-x)BiFe_{0.97}Ga_{0.03}O₃-xBaTiO₃ (abbreviated as BFO-xBT) ceramics were synthesized by conventional solid state reaction method. The effects of BT content on its phase structure, microstructure and electrical properties were investigated. A MPB between rhombohedral (R) and tetragonal (T) phases was found at $x = 0.30$. Moreover, with increase of BT content, the ceramics gradually transform from ferroelectric to relaxor ferroelectric phases. Due to the multiphase coexistence structure at the MPB, the BFO-30BT ceramic exhibited the superior properties with large remnant polarization $P_r=40.8\mu\text{C}/\text{cm}^2$, high dielectric constant $\epsilon_r= 80,000$ at T_C , medium piezoelectric coefficient $d_{33} = 114 \text{ pC/N}$ and $k_p = 0.284$, and high Curie temperature $T_C = 470 \text{ }^\circ\text{C}$. In addition, the temperature-dependent piezoelectric and ferroelectric analysis confirmed that the BFO-BT ceramics possess increasing piezoelectric coefficient d_{33} to 200 pC/N at 300°C, and a large electric-field-induced unipolar strain of 0.5% at 190°C. These superior high-temperature piezoelectric properties indicates the good temperature stability of the BFO-BT based piezoelectric ceramics. These results indicates that the BFO-BT ceramic is a potential lead-free piezoelectric materials for high temperature applications.

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Activation of lead-free (Na,K)NbO₃ piezoceramics using pulsed electric field

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To take advantage of the piezoelectric effect, which describes the ability of a material to transduce electrical into mechanical energy, and vice versa, piezoceramics need prior electrical poling. The poling process is generally carried out at a constant voltage under a direct current (DC)-poling field. Recently, studies have shown that pulsed electric fields can have certain advantages, such as the enhancement of the piezoelectric properties of lead-base materials, over DC-poling fields.^[1, 2] However, these pulsed electric fields can take the form of different waves, such as triangular, sinusoidal, or rectangular, with varying results. In addition, the electric fields can be unipolar, as well as sesquipolar, or bipolar, meaning alternating current (AC)-poling fields. As such, three different types of pulsed electric field using several waveforms were applied to (Na,K)NbO₃ (NKN)-based ceramics. Here, NKN ceramics are a promising candidate for lead-free piezoceramics, as they demonstrate a high Curie temperature and good piezoelectric properties.^[3] In this study, the different effect each pulsed field had on the activation of the piezoelectric properties was compared, and the underlying mechanisms discussed.

0.20 mol% MnO-added (Na_{0.55}K_{0.45})NbO₃ (NKN) ceramics were prepared by a solid-state reaction method. The compacted disks of NKN powder were sintered at 1060°C for 2h. Poling experiments were performed at ambient temperature in a silicon oil bath. Four different waveforms of 3DC and 1AC, as illustrated in Fig. 1(a) - 1(c-2), were applied to the NKN ceramics. The electrical properties were measured using a ferroelectric tester and d_{33} -meter. In order to understand the changes to the electrical properties, characterization of the domain structure was conducted using a scanning electron microscope.

Conventional DC-poling with a poling time of 100 sec resulted in NKN ceramics with a piezoelectric charge coefficient d_{33} of 141.3 pC/N. Equivalent values were achieved by pulsed DC-poling in only 10 sec. On the other hand, a lower value ($d_{33} = 135$ pC/N) was recorded after pulsed AC-poling. As such, the activation of the piezoelectric properties depended on the waveforms of pulsed electric field. A correlation between piezoelectric properties and domain structure was discussed, as well as the viability of pulsed electric fields.

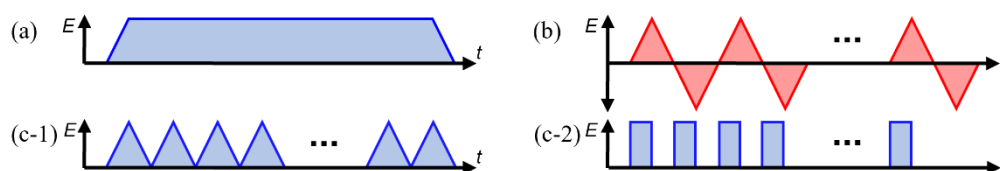


Fig. 1. Schematics of waveforms for (a) conventional DC-poling, (b) triangular pulsed AC-poling, (c-1) triangular pulsed DC-poling, (c-2) rectangular pulsed DC-poling.

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Realization of enhanced strain responses in lead-free (Bi_{0.5}Na_{0.5})TiO₃-based ferroelectric thin films by active domain motions

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Abstract

Lead-free (Bi_{0.5}Na_{0.5})TiO₃-based ferroelectric ceramics and thin films with excellent piezoelectric properties and strain responses possess broad application prospects for precision actuators and transducer applications. In this work, (Bi_{0.5}Na_{0.5})TiO₃-BaTiO₃-NaNbO₃ (BNT-BT-NN) thin films were prepared by sol-gel methods and deposited on Pt/TiO₂/SiO₂/Si (001) substrates. An ultrahigh strain response of 1.7% was obtained in the BNT-BT-NN thin films as the NN substitution of 6%. Low dielectric loss $\tan\delta$ of ~ 0.03 and low leakage current of $\sim 9 \times 10^{-9} \sim 2 \times 10^{-7}$ A/cm² thank to the high quality and well-organized microstructures by optimizing the solution preparation and cautious control of the film deposition process. The BNT-BT-NN samples exhibited a phase coexistence of ferroelectric (FE) phase and relaxor (RE) state in a pseudo-cubic structure. It is believed that the enhanced strain responses are closely related to the reversible phase transition between the RE state and FE phase. Large transient currents related to domain switching measured by PUND method indicated that active domain wall motions contribute to higher dielectric and piezoelectric responses. Further, the intensity of the current peak related to the domain switching in the BNT-BT-NN thin films as the NN substitution of 6% is higher than that of other compositions. The enhanced strain response renders the NN-modified BNT-based thin films appealing for precise actuator and transducer applications.

Preparation of high temperature resistant barium titanate/polyimide flexible nanocomposite energy storage films

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Abstract: Dielectric energy storage capacitors can release stored energy in a very short time (microsecond level) to produce large power pulses. At present, its practical applications placed in high power, high current and high temperature working conditions. Therefore, it is urgent to develop dielectric materials with high energy storage density and high temperature resistance which suitable for high temperature dielectric energy storage capacitors. Here, the two-dimensional barium titanate powder was prepared by combining with solid and hydrothermal method, and the barium titanate/polyimide (BT/PI) composite films were prepared by the diffusion method. When 5% filled at the condition of 150 °C, the energy density of the composite film can reach 0.7 J/cm³, the charging and discharging efficiency can reach 77.1%, and the dielectric constant was stable at about 5.0 within the range of 150 °C.

Keywords: High-temperature energy storage, Polyimide, Barium titanate

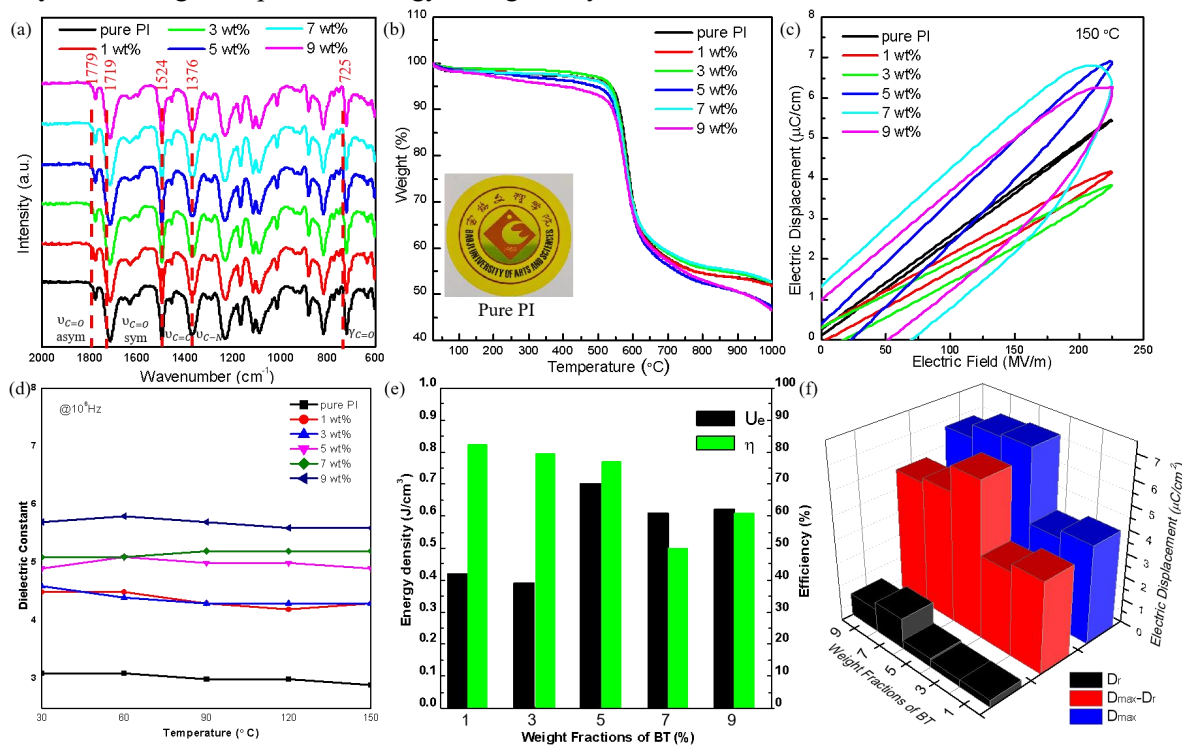


Fig1.(a) The Fourier transform infrared spectra, (b) TG curves, (c) D - E loops at 150 °C, (d) dielectric permittivity on temperature at 10⁶ Hz, (e) Energy density and Efficiency and (f) D_r , D_{max} and $D_{max}-D_r$ of 0, 1, 3, 5, 7 and 9 wt% BT/PI nanocomposite films.

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Change of crystal and microscopic structures of $\text{Ba}_{0.95}\text{Sr}_{0.05}\text{TiO}_3$ ceramics prepared by the combination of reaction sintering and spark plasma sintering

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Controlling the microstructure of ceramics is of great importance to improve their properties. Here, we report the change of crystal and microstructure of $\text{Ba}_{0.95}\text{Sr}_{0.05}\text{TiO}_3$ (BSTO-5) ceramics prepared by the one-step reaction sintering using the system of Spark Plasma Sintering (SPS), in which solid phase reaction and sintering can proceed simultaneously within a very short time under the application of high pressure. It is found that the solid-state reaction between BaTiO_3 and SrTiO_3 can be finished as the sample is heated to 1200 °C at a heating rate of 100 °C /min under a 1 atm N_2 atmosphere . In association with the complete of the reaction, the microstructure is changed sharply, leading to the rapid densification of the BSTO-5 ceramics. By using this approach, we can prepare BSTO-5 ceramic with a relative density higher than 99 % and with an extremely dense and homogeneous microstructure. In addition, the dielectric and ferroelectric properties of the obtained sample have been investigated and will be reported in detail.

Microstructure of AC poled ternary PIN-PMN-PT single crystal after high temperature annealing.

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Ternary system of $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ (PIN-PMN-PT) piezoelectric single crystals (SCs) have been utilized as high-frequency >5 MHz medical array transducers because of their higher coercive field $E_c > 4$ kV/cm and higher phase transition temperature $T_{pc} > 100^\circ\text{C}$ compared to those of binary system PMN-PT SCs. Recently, Yamamoto and Yamashita have reported that enhanced di- and piezoelectric properties are obtained by applying AC polarization (ACP) to SCs in the [001] orientation of the rhombohedral phase compared to conventional DC polarization (DCP), which has attracted worldwide attention. [ref. 1-3]. Many reports have been published on the microstructures of DCP and ACP piezoelectric SCs observed using piezoelectric force microscopy (PFM) and scanning electron microscopy (SEM). The ACP piezoelectric SCs have been reported to show stripe 109° domains of 0.5 to several μm parallel to the electrodes, unlike DCP products. However, there has been no report comparing the microstructure of DCP and ACP SCs once depolarized (DEP) by annealing at 300°C , which was fairly higher than Curie temperature (T_c) of these materials ($T_c = 130\text{-}220^\circ\text{C}$).

We performed DCP and ACP on the 0.24PIN-0.46PMN-0.30PT SC plates with a T_c of 180°C produced by continuous feeding Bridgman method, which provides excellent uniformity of composition and properties, followed by DEP at 300°C for 10 min of annealing. The piezoelectric constants (d_{33}) of the samples after high-temperature annealing was less than 10 pC/N, and no piezoelectric response was observed in the impedance spectra measurements. The microstructures of (a) ACP and (b) ACP-DEP samples observed by SEM were shown in Figure 1. The ACP sample showed stripe like layered structure of 109° domains as reported previously. However, the ACP-DEP sample showed no piezoelectric response but a microstructure with residual striped 109° domains.

This result indicates that stripe 109° domains formed once by ACP still maintained after annealing temperature of 300°C , which is 120°C higher than the T_c even the SC does not show any piezoelectric responses. These results suggest that repeated ACP-DEP on the same piezoelectric SC sample may preserve the past history of the microstructure. Therefore, we have to take care that the microstructures of SC have a past poling history, which may lead to a different judgment of microstructure.

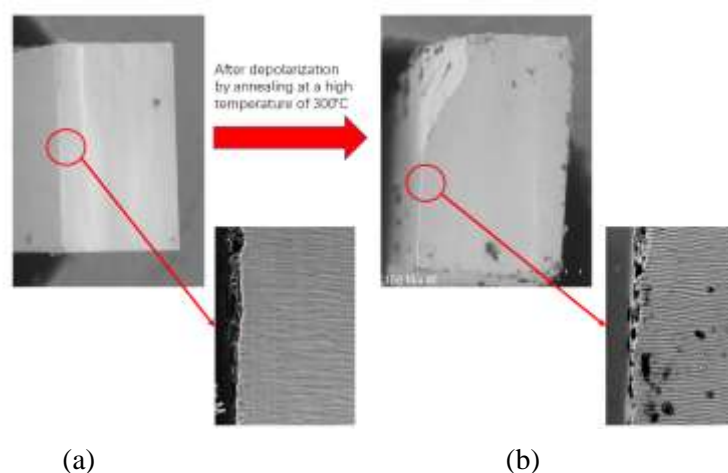


Fig.1 SEM microstructure of (a) ACP and (b) ACP-DEP of PIN-PMN-PT SCs. Note, the ACP-DEP SC showed 109° stripe like domains after 300°C annealing.

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Effects of sputtering conditions on the growth of AlScN thin films and its conduction mechanism

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[Introduction] Recently, $\text{Al}_{1-x}\text{Sc}_x\text{N}$ (AlScN) thin films have attracted much attention due to the increase in piezoelectric constant [1] and the appearance of ferroelectricity [2]. However, (AlScN) thin films have an issue of reliability because the coercive electric field (E_c) is close to the breakdown electric field (E_{bd}) [3][4]. To utilize the ferroelectricity of AlScN thin films in electronic devices, the leakage current should be low enough even near E_{bd} . In this study, the conduction mechanism of AlScN thin films fabricated under different conditions was investigated to discuss the factors affecting the leakage current.

[Experiments and results] AlScN films were deposited by RF magnetron sputtering on 2-inch (111)Si wafer using a $\text{Al}_{0.7}\text{Sc}_{0.3}$ alloy target. The RF power was set to 50 W, and the Ar/ N_2 ratio was varied between 0.09 and 4.0. The substrate temperature and chamber pressure was kept at 300 °C and 0.8 Pa, respectively, during the deposition. Fig. 1 shows the X-ray diffraction patterns of AlScN thin films deposited at various Ar/ N_2 ratios. (0002) oriented AlScN films were grown at the Ar/ N_2 ratio from 0.25 to 1.5. The c-axis lattice constants, calculated from the data in the figure were changed 0.74 %. Fig. 2 shows I-V characteristics of the AlScN film deposited at Ar/ N_2 = 0.25. By increasing the step delay time, which is the delay time for each voltage step, from 0.1 s to 1 s, the leakage current increases. This result indicates that the vacancies in the thin film are filled in a short time.

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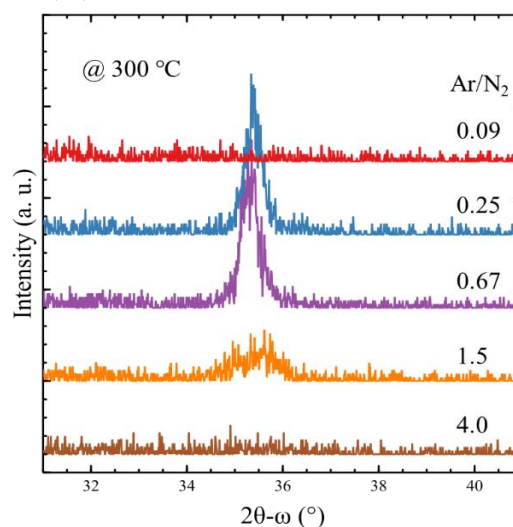


Fig. 1. X-ray diffraction patterns of AlScN films deposited at various Ar/ N_2 ratios.

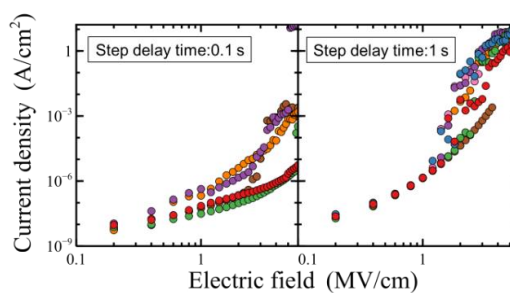


Fig. 2. I-V characteristics of the AlScN film deposited at Ar/ N_2 = 0.25.

High-Performance Neuromorphic Computing Based on Ferroelectric Synapses with Excellent Conductance Linearity and Symmetry

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Artificial synapses can boost neuromorphic computing to overcome the inherent limitations of von Neumann architecture. As a promising memristor candidate, ferroelectric tunnel junctions (FTJ) enable us to successfully emulate spike-timing dependent synapses. However, the nonlinear and asymmetric synaptic weight update under repeated presynaptic stimulation hampers neuromorphic computing by favoring the runaway of synaptic weights during learning.

Here we demonstrate an FTJ (Au/Cr/BaTiO₃/Nb:SrTiO₃) whose conductivity varies linearly and symmetrically by judiciously combining ferroelectric domain switching and oxygen vacancy migration. The artificial neural network based on this FTJ-synapse achieves classification accuracy of 96.7% during supervised learning, which is the closest to the maximum theoretical value of 98% achieved to date. This artificial synapse also demonstrates stable unsupervised learning in a noisy environment for its well-balanced spike timing-dependent plasticity response. Our novel concept of controlling ionic migration in ferroelectric materials paves the way towards highly reliable and reproducible supervised and unsupervised learning strategies.

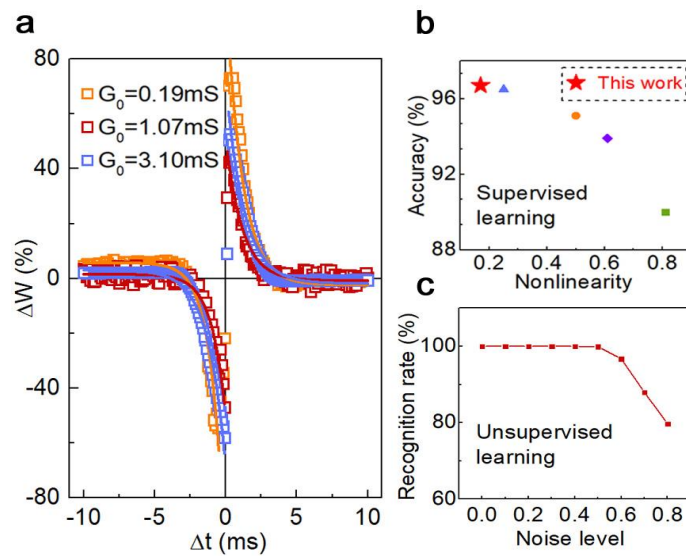


FIG. (a) The balanced STDP is independent of initial states in our devices. (b) The classification accuracy versus nonlinearity of some typical two-terminal artificial synapses. The linear weight update makes the supervised learning classification accuracy in our device the best of all two-terminal devices. (c) The variation of unsupervised learning recognition rate with the noise level. Our device demonstrates that the balanced STDP contributes greatly to the robustness of unsupervised learning.

Low-temperature deposition of transparent $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ thin film on glass substrate with giant piezoelectric response

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$\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT) shows a morphotropic phase boundary (MPB) at the composition of $x=0.47\sim 0.48$, in which high piezoelectricity has been found [1], and its ceramics has been widely used for various devices. Also, PZT thin films deposited on Si substrate have been intensively investigated for the applications in the semiconductor integration devices. For the development of next generation of piezoelectric devices, it is highly desired to deposit PZT film with excellent piezoelectric properties on the transparent glass substrate. However, the issue remained to be solved is how to prepare the PZT film on glass substrate at a deposition temperature lower than its strain point. For the commonly used TFT glass substrate, it is necessary to prepare the PZT film at a low temperature of 550°C . However, the approach for such a low-temperature deposition has not been established.

Here, we report a novel approach to deposit the PZT film on the glass substrate at a low temperature of 550°C , in which the molecularly designed precursors were used. In our approach, a LaNiO_3 (LNO) conducting seed-layer was also introduced to control the orientation growth. Fig. 1 clearly shows that our PZT film deposited at 550°C on glass substrate is transparent and preferably oriented along the [100]/[001] direction. As shown in Fig. 2, our PZT film with MPB composition of $x=0.47$ exhibits a large displacement of 5 nm at an applied voltage of 20 V, which corresponds to an effective piezoelectric response of 250 pm/V that is higher than the reported value of 220 pm/V for the pure PZT ceramics[1]. More importantly, we found that the tetragonal PZT film with $x=0.55$ shows higher piezoelectric response than the PZT film with MPB composition. An extremely large displacement of 10 nm was observed for this film at the same applied voltage of 20 V, which corresponds to a giant piezoelectric response of 500 pm/V. This value is twice of that of PZT film with MPB composition.

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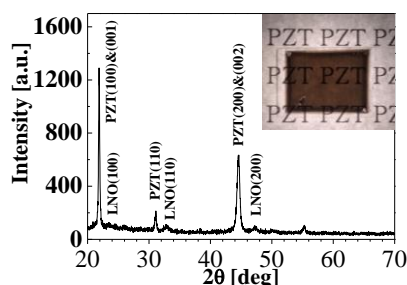


Fig. 1 XRD pattern of the PZT thin film with $x=0.47$. Insert shows the photographic image of the deposited film.

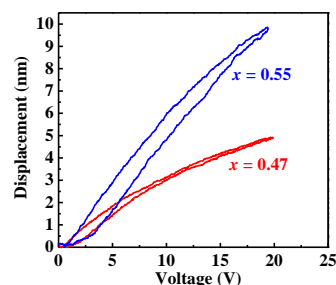


Fig. 2. Displacement curves under the application of the unipolar voltage. The film thickness is estimated to be 800 nm.

Mechanomyography Using Piezoelectric Materials

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Introduction: There are increasing concerns about the utilization of electronic devices in the medical and health fields. Detecting muscle fatigue during exercise is useful for rehabilitation and injury prevention by optimizing exercise intensity. Mechanomyography (MMG) is one of the methods to detect muscle fatigue and a technique that measures minute vibrations during muscle movement using a condenser microphone, accelerometer, and so on [1]. Compared to the widely used surface electromyography (sEMG), MMG is less susceptible to changes in skin condition caused by perspiration and is expected to be used under intense exercise and in daily life. However, due to the large noise caused by motion artifacts, it has not yet been realized for these applications. In this work, we fabricated a sensor using organic piezoelectric material and investigated a way to measure muscle movement.

Measurement and result: Fig. 1 shows the structure of a fabricated sensor. P(VDF-TrFE-CFE) sheet with a thickness of 25 μm , manufactured by PolyK, was used for the sensor. Pt electrodes with a thickness of about 40 nm were deposited on both sides of the P(VDF-TrFE-CFE) sheet by sputtering. The frequency dependence of the dielectric constant and loss shown in Fig. 2 indicates that the capacitor structure is formed as expected.

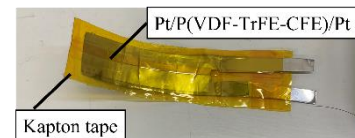


Fig. 1 The structure of the vibration sensor.

Then, comparative experiments of sEMG and MMG were conducted. Commercially available electrodes (Ag/AgCl) were placed parallel to the muscle fibers as shown in Fig. 3 and the MMG sensor was also placed between them. The interface between the MMG sensor and the skin was wetted with water to facilitate the transmission of the muscle sound. Fig. 4 shows the measurement results. The peaks of the sEMG and MMG signals appear synchronously, which indicates that MMG can be measured with the fabricated sensor. It appears that the large SNR of MMG is caused by poor contact with the skin.

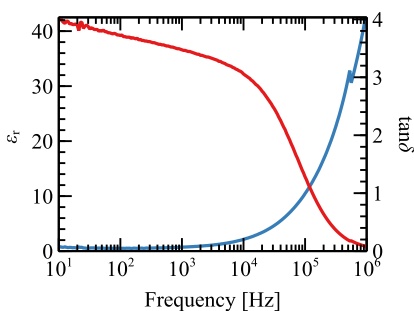


Fig. 2 Dielectric constant and $\tan\delta$ as a function of frequency.

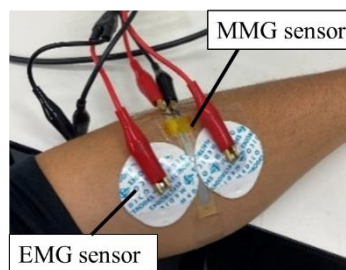


Fig. 3 Placement of sEMG and MMG sensors on the arm.

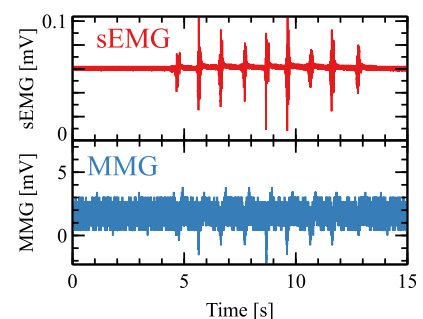


Fig. 4 sEMG and MMG signals.

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Unique Gas Sensing Performance of Sol-gel Derived BiFeO₃ Nanoparticles

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Perovskite BiFeO₃ (BFO) oxides, a kind of ferroelectric semiconductors with rich physical and chemical properties, have been regarded as promising sensing materials for the toxic and harmful gas detection. In this talk, BFO nanoparticles and tourmaline@BFO composites were prepared by a facile sol-gel method. Micro-Electro-Mechanical System (MEMS) gas sensors were fabricated by using BFO based materials. The corona poling treatment has great effects on the gas sensing behavior by regulating the polarization field and carrier movements of BFO nanoparticles. The BFO-P4 (poled under 4 kV) sensor exhibits a high response value of 4.8 for H₂S with concentration of 1200 ppb at 220°C, achieving a detection limit of 10 ppb and a short response/recovery time of only 3s/7s respectively. In addition, tourmaline@BFO composites with the growth of nano-sized BFO on the tourmaline laminar exhibit the temperature-dependent dual selectivity for C₃H₆O and H₂S at 150 and 210 °C respectively. Our results indicate that BFO based sensors with unique gas sensing performances have great potentials in detection of polar H₂S and C₃H₆O gases.

Key words: BiFeO₃ nanoparticles; tourmaline@BiFeO₃ composites, corona poling; MEMS gas sensor; H₂S and C₃H₆O

Ferroelectric and piezoelectric properties of c-axis oriented $\text{PbZr}_{0.53}\text{Ti}_{0.47}\text{O}_3$ thin film deposited on SUS substrate with large coefficient of thermal expansion

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$\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT) ceramics and thin films with composition near the morphotropic phase boundary (MPB, $x=0.47-0.48$) have been widely applied due to their excellent ferroelectric and piezoelectric properties. However, there is a lack of understanding of its intrinsic physical properties owing to the difficulty in the growth of single crystal since it was discovered several decades ago. Here, we report the ferroelectric and piezoelectric of polar-axis-oriented PZT thin film with the MPB composition ($x=0.47$). The PZT film was deposited on SUS substrate that has a 1.5-time coefficient of thermal expansion of PZT. Such larger coefficient of thermal expansion allows to apply an in-plane compressive stress to the PZT film as cooling down from the firing temperature to room temperature, leading to the c-axis orientated growth of PZT film with the help of the seed layer of a-axis oriented LaNiO_3 (LNO) electrode. Our PZT precursor was prepared by a partial hydrolysis method and had a high degree of homogeneity of B-sites. As evident from the x-ray diffraction pattern shown in Fig. 1, the c(polar)-axis oriented PZT film has been successfully prepared on the SUS substrate. More importantly, the remanent polarization of our PZT film shows nearly the same value of the saturation polarization, indicating that all ferroelectric domains were aligned along the same direction under the application of a high electric field. This is further confirmed by the displacement-field loop, in which a pure linear piezoelectric response was observed when the domains were nearly aligned along the field direction for $E > E_c$. The piezoelectric constant d_{33} of PZT along the polar axis was then calculated from this linear relationship between the displacement and the applied voltage and was estimated to be 40~49 pm/V from several samples. Our experimental result is in good agreement with the value of 50~55 pC/N predicted from first principles calculations for PZT single crystal.^[1] Our finding indicates that the large piezoelectric response observed in PZT polycrystalline is essentially dominated by the domain effects.

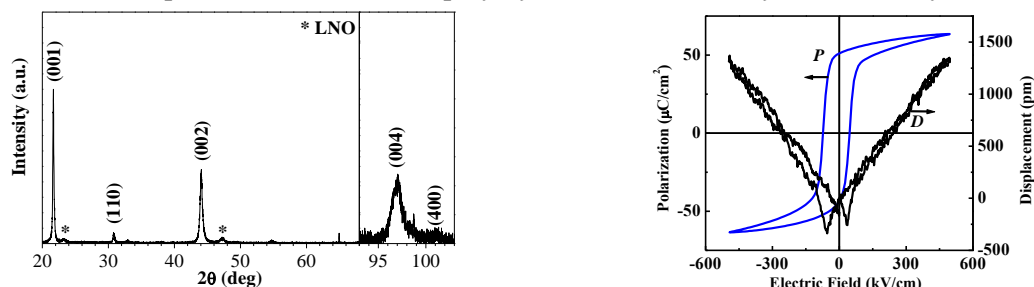


Fig.1 XRD patterns of PZT film with $x=0.47$. Fig.2 P - E and displacement loops of PZT film with $x=0.47$.

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Superior energy storage properties in NaNbO₃-based ceramics via synergistically optimizing domain and band structures

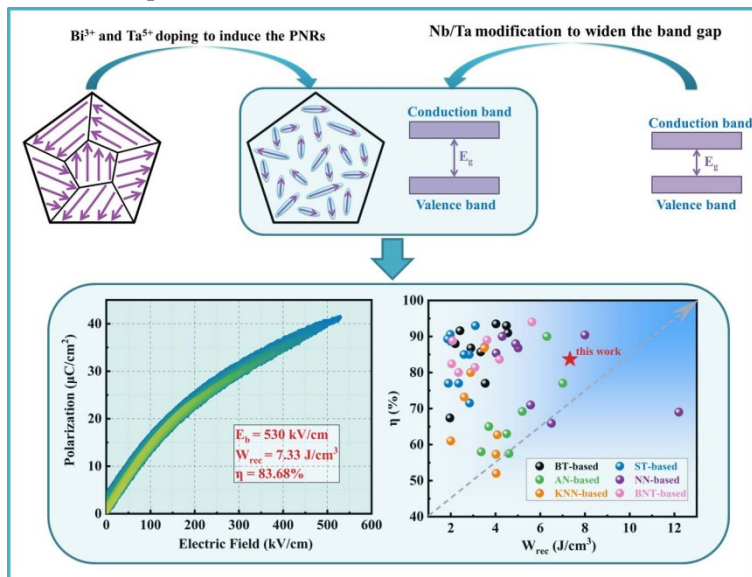
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Environmental-friendly ceramic capacitors with outstanding energy storage properties (ESP) are greatly desired for the advanced pulsed power systems. However, it is still a great challenge to develop lead-free dielectric materials with simultaneous excellent recoverable energy storage density (W_{rec}) and energy storage efficiency (η). In the present work, a synergistic optimization strategy with regard to ferroelectric domain and band structure is applied to NaNbO₃ (NN)-based ceramics, where the introduction of Bi³⁺ induces the elongated ferroelectric P-E loops due to nanodomains formation, and the further modification of Ta doping content remarkably reduces the grain size and widens the band gap (E_g), leading to a high breakdown strength. As a result, the involved Na_{0.7}Bi_{0.1}Nb_{0.9}Ta_{0.1}O₃ ceramics exhibit excellent comprehensive ESP ($W_{rec} = 7.33 \text{ J/cm}^3$, $\eta = 83.68\%$, $E_b = 530 \text{ kV/cm}$) and good stability. Moreover, ultrafast discharge time of 60 ns and high power density of 320.21 MW/cm³ are also achieved. The Na_{0.7}Bi_{0.1}Nb_{0.9}Ta_{0.1}O₃ ceramics with uncomplicated chemical composition and prominent properties demonstrate promising applications to pulsed power system and the synergistic optimization strategy in this study offers an important clue for future lead-free ceramic capacitors.



Extremely large strain response under low driving electric fields in lead-based textured piezoelectric ceramics

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Precision positioning technology is one of the key technology to achieve precision manufacturing, precision measurement and precision drive, and it has been receiving great attentions and widely used in optical engineering, aerospace, semiconductor industry and many other high-tech fields. As the key part of precision positioning system, micro-displacement actuator play an important role in sensitivity and resolution of precision positioning system. In order to achieve large driving force and high-precision displacement of micro-displacement actuator, it is particularly necessary for piezoelectric ceramics to possess a high strain value and low strain hysteresis under small driving electric fields. Up to date, it remains grand challenge for piezoelectric ceramics to simultaneously possess large strain response and low strain hysteresis under low driving electric fields. At present, texture technique, which can make ceramic grains oriented along specific crystallographic orientation, is considered one of the most promising means to significantly improve the strain response of piezoelectric ceramics. In this work, we adopted a combination strategy of composition design and texture technique to develop a novel textured ceramics to address this challenge. In this paper, 1%Sm-PMN-30PT textured ceramics with highly [001]_c orientation was successfully manufactured and an ultrahigh strain response of 0.27 % and a low strain hysteresis of 15 % at 2 kV/mm were obtained concurrently.

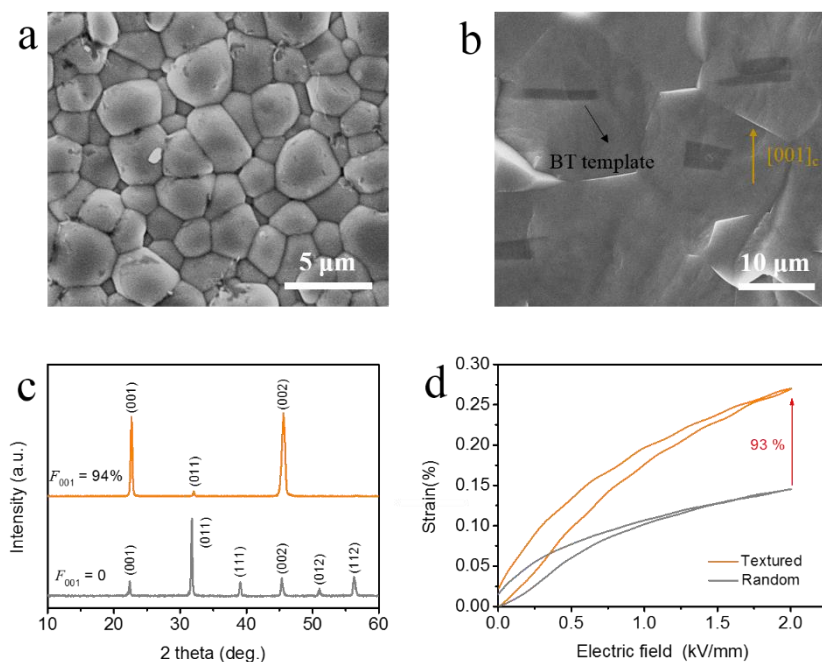


Fig. 1. Structural and strain characterizations of 1%Sm-PMN-30PT ceramics. (a) Surface SEM image of random ceramics. (b) Fracture surface SEM images of textured ceramics. (c) XRD patterns of random and textured ceramics. (d) Unipolar strain curves of random and textured ceramics.

Experimental approach to residual hydrogen content in silicon substrates by proton beam writing

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Water lithography, electron beam induced reaction, inkjet printing, screen printing, three-dimensional printing, etc. have been actively investigated as microfabrication techniques for piezoelectric materials. Among them, we think that the focused proton beam writing (PBW) is very effective for microfabrication of piezoelectric materials containing heavy elements. However, in our previous report, it has been confirmed that irradiated protons go through piezoelectric films to reach the substrate by Monte Carlo simulation results.¹⁻³ In semiconductor technology, hydrogen implantation is used to manufacture Silicon-On-Insulator (SOI) wafers, formation of porous silicon, and increase the resistance of silicon substrates.⁴⁻⁶ This suggests that the protons remaining in the substrate affect the characteristics of the fabricated device. Therefore, in this study, the residual amount of irradiated protons to the silicon substrate was estimated by the infrared (IR) absorption method.

A focused proton beam lithography system (KOBELCO, MB-S1000) with an acceleration voltage of 1 MV was used to prepare the measurement silicon samples. In this study, silicon with a resistivity of 5 ohm-cm was used as the proton-irradiated substrate. The IR absorption spectra were measured using FTIR (JASCO, FT/IR-4100) in a dry nitrogen atmosphere.

Figure shows the IR spectra at various proton doses. As can be seen from the figure, absorption due to the Si-H bonds could not be observed in the irradiation range up to $300 \mu\text{C}/\text{cm}^2$. Monte Carlo simulation results indicate that most of the irradiated protons are distributed only within $2 \mu\text{m}$ in the depth direction. In this region, the maximum hydrogen concentration is calculated to be 0.02 at%. However, it is possible that the detection limit has not been reached. Therefore, we will continue to evaluate the amount of residual hydrogen in the substrate by further increasing the proton dose.

Acknowledgements

The authors would like to thank Prof. H. Nishikawa and Mr. K. Kawamura for their cooperation in sample fabrication.

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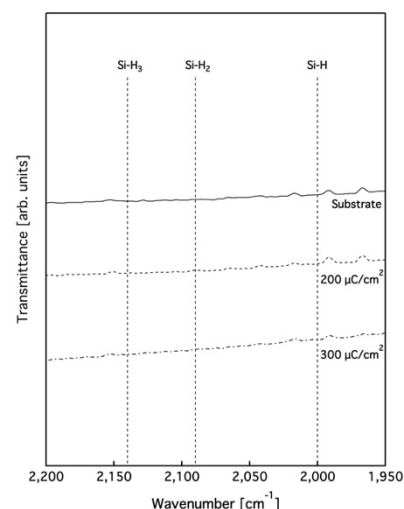


Fig. IR spectra of silicon substrates with various proton doses. The spectrum has not been normalized.

Polarization Rotation Control Domain Dynamic Response Modulates Piezoelectric Properties of Lead-Free Thin Films

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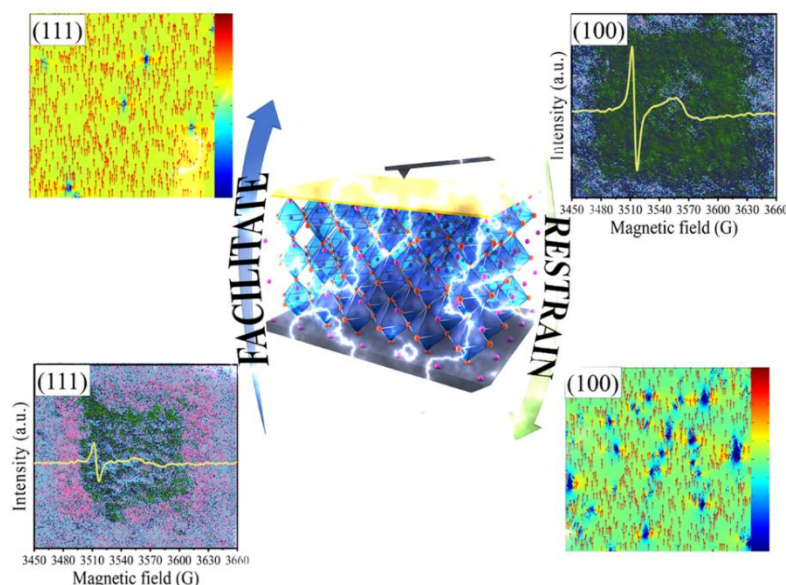
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Domain orientation engineering regulates the polarization direction of thin film and facilitates the enhancement of piezoelectric properties. A change in polarization direction implies a structural change in the piezoelectric thin film. In this work, some unexpected results have been obtained by preparing three orientations of $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ -based lead-free piezoelectric thin films to amplify this structural change. In distinction to the conventional perception, the piezoelectric properties decrease with increasing polarization direction. This anomaly is influenced by the concentration of structural defects, and rotation of the polarization direction may induce a change in the electronic structure of the material and hence in the concentration of structural defects. Ultimately, the suppression of the domain dynamics response by increasing structural defects leads to the worst piezoelectric properties for thin films with the traditionally perceived optimal (100) orientation. The revelation of this anomaly has implications for the design of the next generation of high-performance piezoelectric thin films.



High Energy Storage Performance and Large Electrocaloric Response in $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ Thin Films

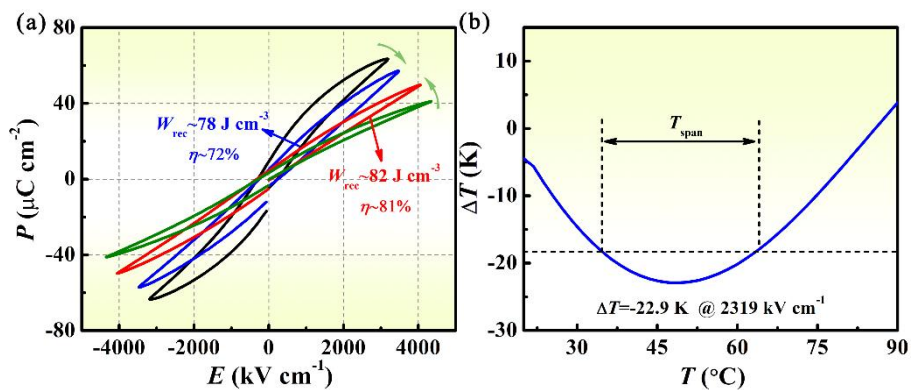
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With regard to the global energy crisis and environmental pollution, ferroelectric thin films with unique polarization behavior have garnered considerable attention for energy storage and electrocaloric refrigeration. Herein, a series of $(1-x)\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}x\text{Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ ($x = 0.3\text{-}0.9$; $(1-x)\text{BNT-xBZT}$) films was fabricated on Pt(111)/Ti/SiO₂/Si substrates. Incorporating BZT can tune the polarization behavior and phase transition temperature of BNT. In this work, high energy storage performances of $W_{\text{rec}} \approx 78 \text{ J cm}^{-3}$ with $\eta \approx 72\%$ and $W_{\text{rec}} \approx 82 \text{ J cm}^{-3}$ with $\eta \approx 81\%$ were observed in the 0.5BNT–0.5BZT and 0.3BNT–0.7BZT films, respectively. Both the capacitors have wide frequency and operating temperature ranges (0.5–20 kHz, 20 °C–200 °C), robust fatigue endurance (10^7 cycles), and ultrafast discharge speed ($<0.4 \mu\text{s}$). Meanwhile, the 0.5BNT–0.5BZT film exhibited a high EC effect near room temperature ($\Delta T \approx -22.9 \text{ K}$, $\Delta S \approx 33.4 \text{ J K}^{-1} \text{ kg}^{-1}$). These remarkable performances signify that the $(1-x)\text{BNT-xBZT}$ system is a promising multifunctional electronic material for energy storage and solid-state cooling applications.



Enhanced stabilization of oxygen vacancies on the bipolar fatigue resistance of PNN-PZT-based ceramics

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The MPB structure PZN-PNN-PZT ceramics were prepared by Mn ions doping and the bipolar fatigue behavior of samples was investigated at both $0.6 E_c$ and $2 E_c$ fatigue fields. Enhanced bipolar fatigue resistance is found by 0.1wt% Mn doping and the remnant polarization P_r only reduces to 0.3% ($0.6 E_c$) and 5.8% ($2 E_c$) of the initial value after 10^6 fatigue cycling. In this work, it is implied that in contrast to the undoped sample, little oxygen vacancy with a weak pinning effect is advantageous to fatigue resistance. These oxygen vacancies hinder crack propagation and stabilize the domain structure during repeated electric cycling. The successive increase of oxygen vacancy leads to the severe pinning effect and degradation of polarization. The influence of oxygen vacancy on fatigue behavior depends on the amplitude of the fatigue field. The characteristics of ceramics in different fatigue fields can be attributed to the pinning effect, mechanical damage, or coexistence of both for different Mn content. This finding is significant to ceramics with MPB structure and has a guideline for the hard material applied in electric field cycling.

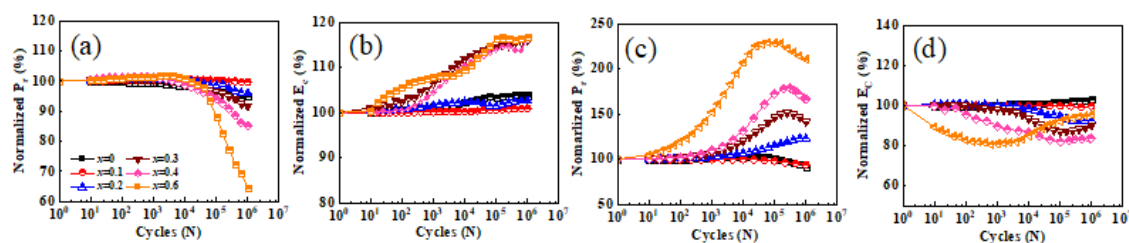


Figure 1. The normalized P_r and E_c of Mn-doped PZN-PNN-PZT ceramics as a function of (a-b) $0.6 E_c$, (c-d) $2 E_c$ bipolar fatigue cycles.

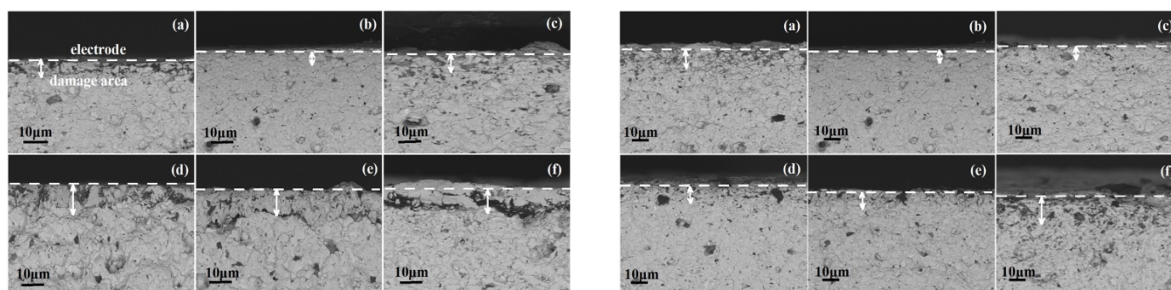


Figure 2 and Figure 3. The SEM images for cross sections of Mn-doped PZN-PNN-PZT ceramics fatigued at $0.6 E_c$ and $2 E_c$. (a) $x=0$; (b) $x=0.1$; (c) $x=0.2$; (d) $x=0.3$; (e) $x=0.4$; (f) $x=0.6$.

Preparation of Tetragonal BaTiO₃ by Molten Salt Synthesis Process

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Abstract: In this work, BaTiO₃ powders were synthesized by molten-salt synthesis process using TiO₂, Ba(OH)₂, and the eutectic salts (NaCl, KCl) as the raw materials. A systematic study is investigated that it indicates the effects of calcination temperature, time as well as the eutectic salts such as NaCl, KCl and NaCl-KCl are related to Ba(OH)₂-TiO₂ on the microstructures of BaTiO₃ powders. The results showed that all powders crystallized with tetragonal perovskite structure, it further confirmed through X-ray diffraction pattern and Raman spectra. Tetragonal BaTiO₃ were prepared by molten-salt synthesis process of reacting TiO₂, Ba(OH)₂ and KCl at 770 °C for 2 h. The prepared BaTiO₃ powders exist characteristic tetragonal splitting of 002/200 in XRD peaks. Meanwhile, it also supports the tetragonal symmetry ($c/a = 1.0139$). The present results provide a generalized methodology for large-scale syntheses of other perovskite transitional metal oxide powders with less energy consumption.

Key Words: Tetragonal BaTiO₃, Eutectic salts, Molten-salt synthesis process

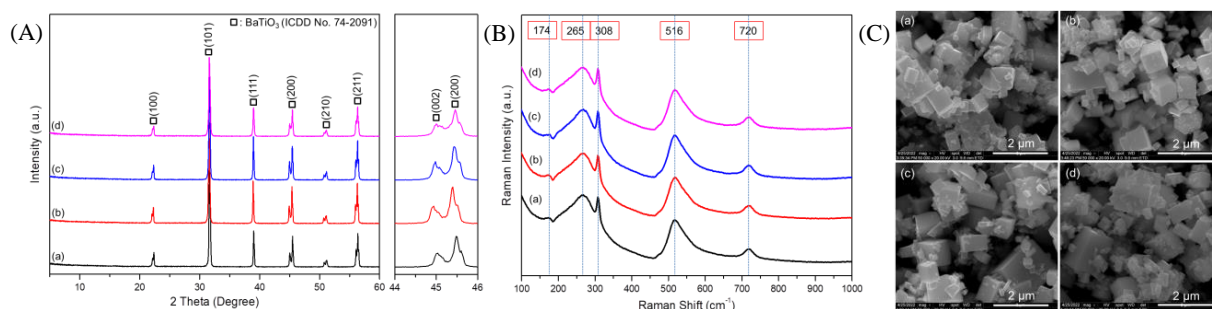


Fig.1 (A) XRD patterns, (B) Raman spectra and (C) SEM images of the BaTiO₃ powders prepared by molten-salt synthesis method of reacting TiO₂ and Ba(OH)₂ at 770 °C for 2 h with (a) 20 %, (b) 30 %, (c) 40 %, (d) 50 % KCl contents.

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Energy storage properties of antiferroelectric PbZrO₃ thin films prepared by chemical solution deposition method

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PbZrO₃ (PZ) is a typical antiferroelectric material and is expected to be a high-density energy storage material. Its energy storage properties have been extensively studied both for the bulk ceramic and thin film. Generally, thin films exhibit a higher energy storage density (W_{rec}) than its ceramics, and the high energy storage density of 19.4 J/cm³ has been reported for the PZ thin film^[1]. However, further improvement of its properties is required for the applications. Here, we report the enhancement of the energy storage density of PZ thin film prepared by chemical solution deposition (CSD) method from the molecularly designed precursors. In the study, we have developed a novel process to prepare the precursors using partial hydrolysis, which allows us to improve the crystalline quality of the PZ film. The PZ film was deposited on a Si substrate with a buffer layer of LaNiO₃ (LNO). The LNO buffer layer, which acts as the bottom electrode for the electrical measurements, was successfully controlled into the [100]-axis orientation, leading to the local epitaxial growth of PZ film. It is found that the films prepared Molecular-design of the precursor solution by partial hydrolysis also resulted in the thicker film thickness per layer than those prepared without partial hydrolysis, because of the higher degree of polymerization and higher homogeneity of the precursor solution. The PZ film with partial hydrolysis shows a larger value of energy storage density of 23.1 J/cm³ that is higher than the reported one^[1]. Moreover, this value is 1.4 times higher than that of the PZ film without using the partial hydrolysis. It seems that the molecularly designed precursors and the LNO buffer layers are of great importance to improve the energy storage properties of PZ film.

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3D Printing of Piezoelectric Biological Scaffolds for Bone Tissue Engineering

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Abstract: Numerous studies have shown that the process of bone repair and regeneration requires a variety of physiological cues, including biological, electrical and mechanical cues. Piezoelectric materials can regulate the physiological micro-environment by changing the mechanical and electrical stimulation environment, which shows excellent potential in bone repair. However, the traditional processing method cannot satisfy the diversified structure of piezoelectric biological scaffolds. 3D printing technology has been introduced into bone tissue engineering due to its high efficiency, high resolution, low cost, efficient use of materials and the ability to manufacture complex structures, it bringing a new dawn for bone tissue repair. Here, the ceramic paste for printing was prepared by mixing the nano-BaTiO₃ (BT) particles, commercial nano-Ca₁₀(PO₄)₆OH (HA) and photosensitive resin. Then the porous interconnecting ceramic bracket and ceramic cylinder are printed by digital light processing technology. The compact structure of the ceramics was obtained after calcination, and the dielectric constant ϵ_r reaches 2733 when 90 wt% BaTiO₃ doped ceramics. DLP printing technology can prepare high precision ceramic supports.

Key Words: 3D printing technology, BaTiO₃, Ca₁₀(PO₄)₆OH, Piezoelectric scaffolds.

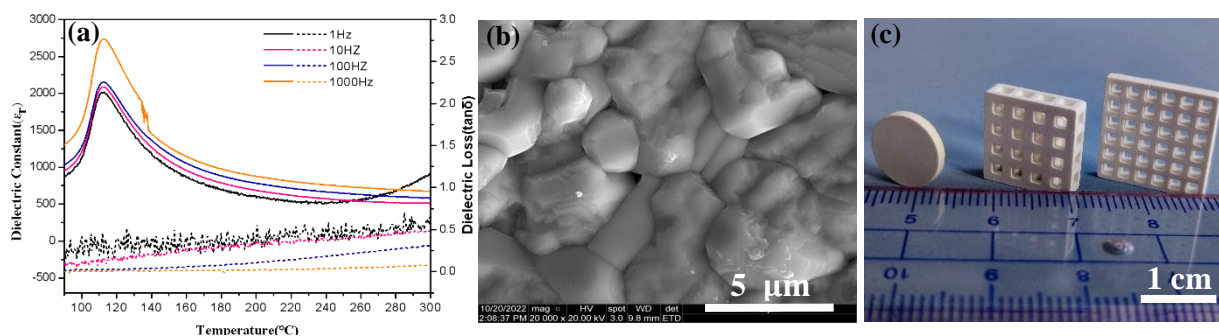


Fig.1 (a) Temperature dependence of dielectric constant and dielectric loss, (b) SEM image of 90wt%BT/HA sample after calcination, (c) Different types of piezoelectric scaffolds for 3D printing.

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Flexible Piezoelectric Nanogenerators Based on Two-dimensional Barium Titanate Nanosheets and Polymer Polylactic acid

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Abstract: With the increasing demand for green energy, piezoelectric nanogenerators (PNG) obtain extensive attention in energy harvesting. In this work, high-performance PNG were successfully fabricated from the composite of BaTiO₃ (BT) (Fig.1b) and polylactic acid (PLA) via spin-coating process. Among, high aspect ratio BT nanoparticle with 2D plate-like morphology synthesized by the solvothermal method which significantly enhanced the piezoelectric properties of the BT/PLA nanocomposite (Fig.1e). The piezoelectric properties of PNG enhanced with the increasing of doping BT. The PNG with 6 wt% BT achieved a maximum output voltage of 11.97 V (Fig.1f) and short-circuit current of 560 nA (Fig.1e). This study shows potential applications of flexible 2D BT/PLA nanocomposite in human motion monitoring, smart skin, and soft robotics. Likewise, it provides an ingenious idea for the disposal of wasted PNG devices and the recycle of piezoelectric fillers.

Keywords: Piezoelectric nanogenerators, BaTiO₃, Polylactic acid.

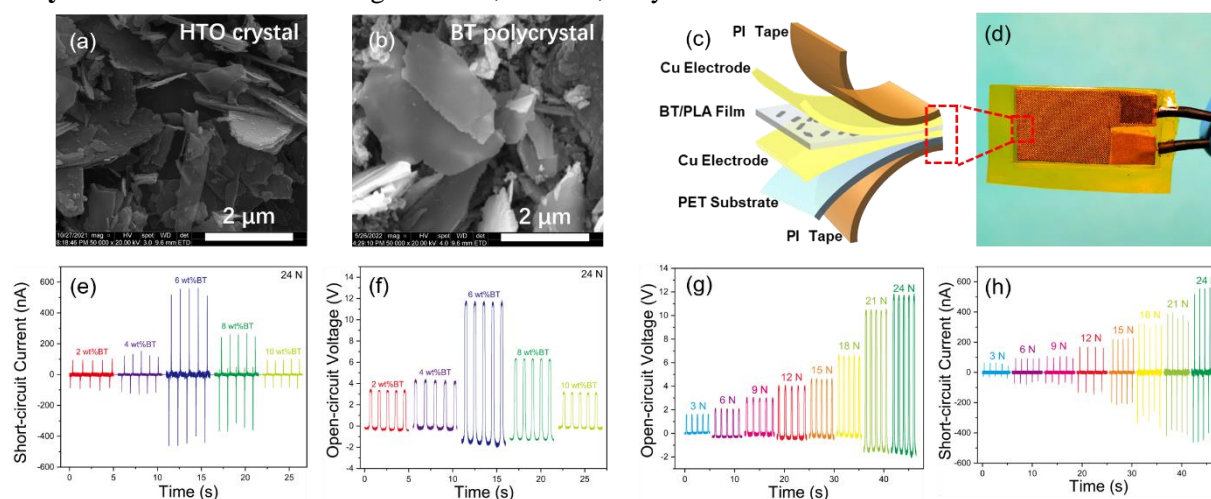


Fig.1 SEM images of (a) HTO and (b) BT crystals; (c) Schematic diagram of PNG structure and (d) Digital photo of flexible BT/PLA nanocomposite. (e) output voltages and (f) currents of BT/PLA PNG with BT contents of 0, 2, 4, 6, 8 and 10 wt%, respectively. (g) Open circuit voltages and (h) short-circuit currents of 6 wt % BT/PLA nanocomposite films under different pressures.

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Energy storage of BT/PVP/PVDF nanocomposite films with multilayer structure

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Abstract: Dielectric capacitors are widely used in modern electronic technology. They have the advantages of ultra-fast charge and discharge rates and ultra-high power density. In order to meet the increasing demand for miniaturization of electronic products, dielectric capacitors with high energy storage performance have been extensively studied. Barium titanate/polyvinylidene fluoride (BT/PVDF) based nanocomposite membrane is a kind of energy storage composite with excellent physical and chemical properties, electrical properties and energy density. How to improve the structural defects of inorganic nanomaterials and the interface polarization between materials are a serious problem at present. Further improving the energy storage performance of composite materials is closely related to the capacitor structure, that is, reasonable structural design is needed. The current research is expected to achieve the multi-scale engineering ceramic structures. Therefore, BT/PVP/PVDF multilayer ceramic capacitors will be an important research direction in the future.

Key Words: BaTiO₃, Energy storage, Dielectric capacitor.

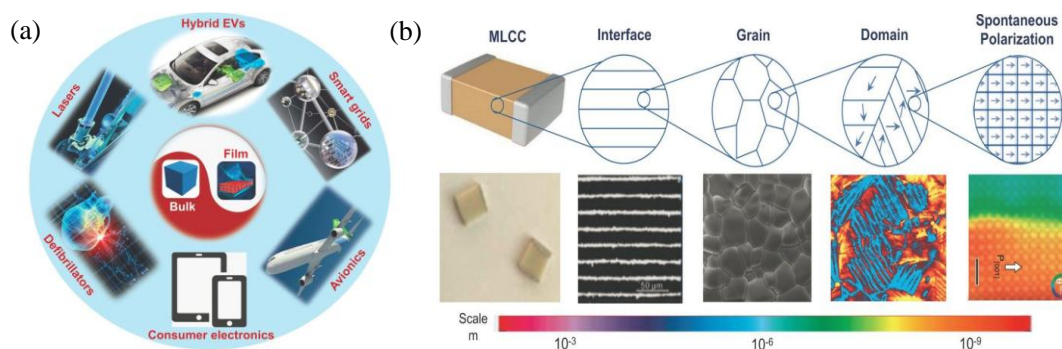


Fig.1 (a) Representative emerging applications of dielectric capacitors, (b) Schematic illustrations (top panel) and direct observations (bottom panel) of multiscale structures, including interface, grain, domain, and spontaneous polarization, for typical ferroelectric ceramic-based MLCCs.

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Fabrication of (Ba,Sr)TiO₃ epitaxial thin films on mica substrate toward flexible electronics

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Flexible devices are capable of attaching to curved surfaces and are expected to be applied especially in the medical and healthcare fields. While the piezoelectric effect is often used in strain sensors or vibration power generator, the flexoelectric effect is also effective for flexible devices with nanostructures. The flexoelectric effect is an electrical polarization phenomenon caused by a strain gradient and is observed in all dielectrics. This phenomenon is especially prominent in perovskite oxides, and it is known that (Ba,Sr)TiO₃ (BST) exhibits a high flexoelectric coefficient near the ferroelectric phase transition temperature. In this study, BST thin films were fabricated on a mica substrate, which is known as a flexible substrate in order to demonstrate flexoelectric flexible devices.

Pulsed laser deposition (PLD) method was used to fabricate the thin films. To promote the crystal growth of the perovskite phase, a seeding layer of CoFe₂O₄ (CFO) was first deposited on (001) mica substrate. SrRuO₃ (SRO) layer was deposited as the bottom electrode, and then Ba_{0.5}Sr_{0.5}TiO₃ thin film was deposited on the SRO layer.

Figure 1 shows the photograph of BST/SRO/CFO/mica flexible structure with a lateral size of 12 × 12 mm². The mica substrate was peeled off to a thickness of about 20 μm. Out-of-plane XRD results showed that only mica (00*l*) and perovskite (111) diffraction peaks were detected, indicating the BST thin film exhibits high crystallinity without any secondary phases. Furthermore, the φ -scans of BST (002), and mica (202) were performed to determine the in-plane orientation relationship. Based on the XRD results, the epitaxial relationship of the heterostructure can be determined as (111) BST / (001) mica. Figure 2 indicates the dielectric property of BST thin film. The relative permittivities of the thin films were around 200 and the dielectric losses were less than 0.1. Flexible structure consisting of a high-quality dielectric film could be fabricated.



Fig.1 Photograph of BST/SRO/CFO/mica flexible structure.

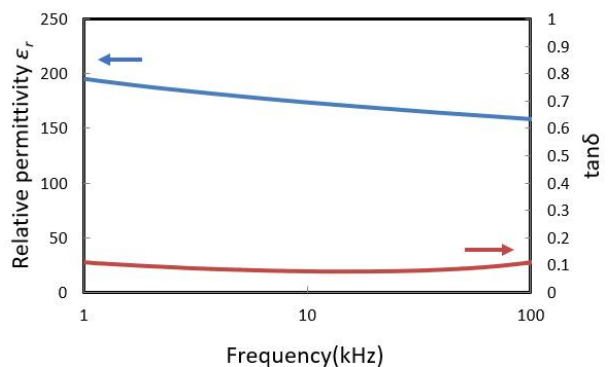


Fig.2 Dielectric property of Ba_{0.5}Sr_{0.5}TiO₃ thin film on mica substrate.

About the Best Poster Presentation Award

The best poster presentation award certificates and some memorial goods for student poster presentations will be given from the Dielectric Society Japan (DSJ) by the Poster Presentation Award Judging Committee.

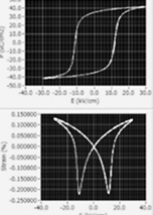
The results will be shown in the CJFMA14 website.



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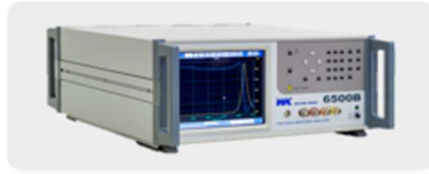
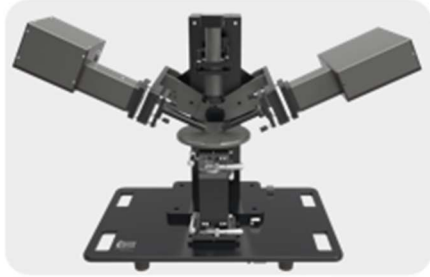


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We are looking for contributors on, but not limited to, the following topics:

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(single crystal, ceramics, amorphous, thin film, polymer, liquid crystal, composite, etc.)

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Key Dates

Short Abstract Submission: January 31, 2023

Extended Abstract Submission: April 10, 2023

Manuscript Submission: May 27, 2023
(JJAP Special Issue)

Venue

60th Anniversary Hall,
Kyoto Institute of Technology, Japan

Fees (Japanese Yen)

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DESJ Student Member *1	5,000	6,000
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