Centre for Interdisciplinary Research on Micro-Nano Methods

ACTIVITY REPORT

April 2022 - March 2023

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Professor & Director
Centre for Interdisciplinary Research on Micro-Nano Methods
IIS University of Tokyo

ACTIVITY REPORT

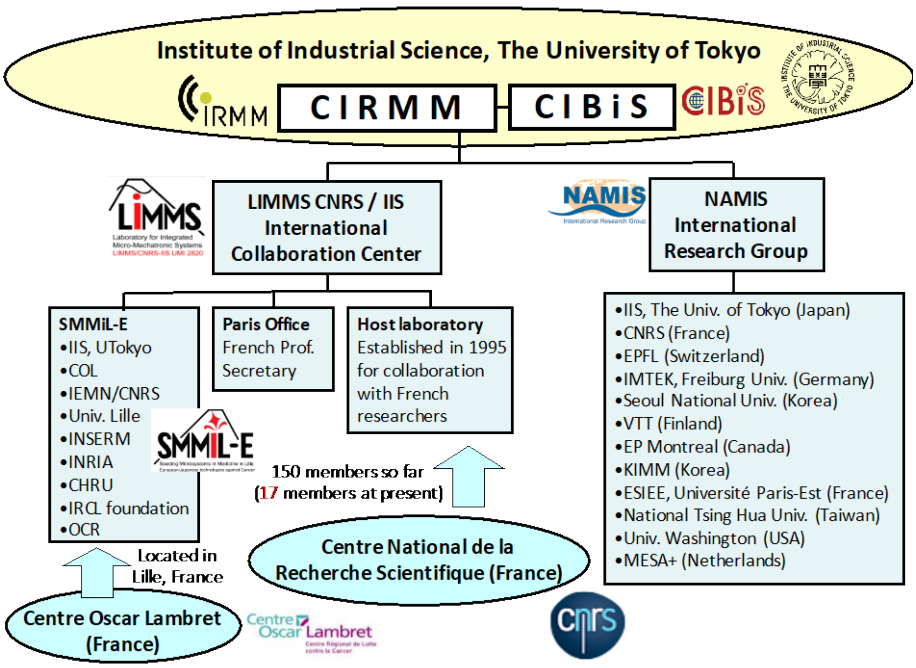
CONTENT

Centre for Interdisciplinary Research on Micro-Nano Methods (CIRMM)

1-1. What is CIRMM

The CIRMM was established in 2000 to facilitate international collaboration on micromechatronics research, which is the study of micro miniature systems composed of mechanical, electrical, optical and bio/chemical devices. The prospect is to obtain high-performance multi-functional devices using heterogeneous integrated process over scale and materials. The first phase of CIRMM (April 2000 – March 2010) finished successfully. The second phase started for a 6-year mandate in April 2010 and come to an end in March 2016. The third phase has started in April 2016 for another 5 years. The name of the center becomes Centre for Interdisciplinary Research on Micronano Methods. Professors conducting biomedical MEMS research in CIRMM created a new center, named "Center for International Research on Integrative Biomedical Systems (CIBiS)" jointly with other professors working on bioengineering on April 1, 2015. Those two centers share the same technological base, i.e. MEMS, and will work together to continue the international activities of CIRMM.

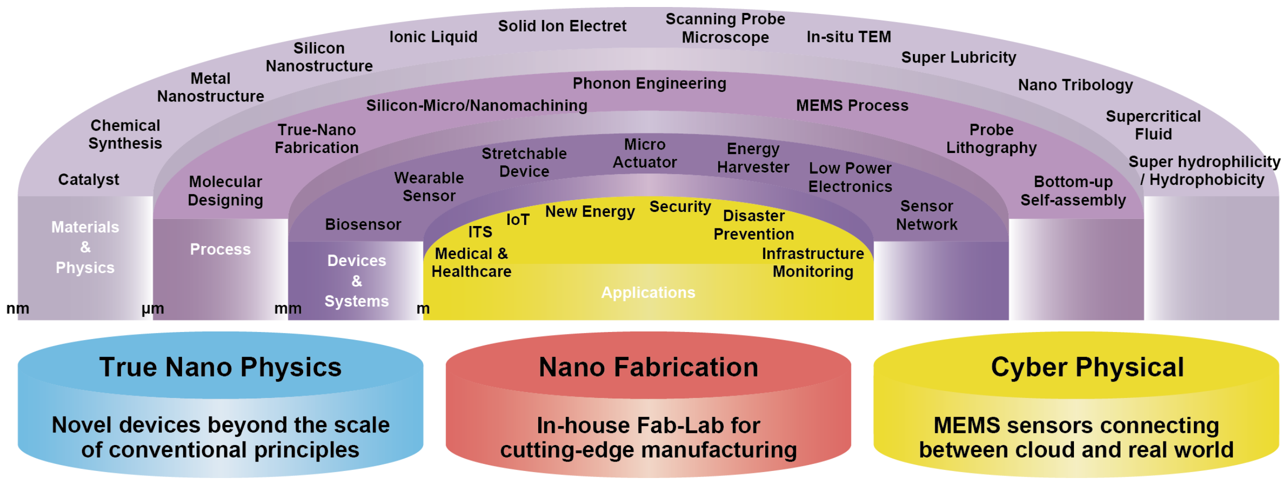
The international activities of CIRMM date back to 1995 with the creation of Laboratory for Integrated Micro-Mechatronic Systems (LIMMS); the joint research laboratory between Institute of Industrial Science(IIS) in the University of Tokyo and Institut des sciences de l'ingènierie et des systèmes (INSIS) in Centre National de la Recherche Scientifique (CNRS). After 9 years of operation, in 2004, the LIMMS became the first Unité Mixte Internationale de Recherche (UMI 2820) of CNRS situated in Asia. International research networks on nano and micro systems (NAMIS) were organized in 2005. Using the network, LIMMS obtained an EU FP-7 project (EUJO-Limms) in 2011 to extend its activities to other European countries, namely Germany, Finland, Switzerland, and Netherlands. On July 16, 2014, SMMiL-E* laboratory, mirror site of LIMMS was founded at Centre Oscar Lambret in Lille, in North France. Bio-MEMS technology of CIRMM is applied to cancer research in SMMiL-E. IIS/UTokyo Bureau for European Collaboration (IBEC) is open in July, 2015 in Lille for both supporting the SMMiL-E project and strengthening EU relation.



CIRMM Organization

SMMiL-E stands for Seeding Microsystems in Medicine in Lille - European Japanese Technologies against Cancer; this program aims at technology transfer of state-of-the-art bioMEMS research performed in IIS/The University of Tokyo and LIMMS/CNRS-IIS towards the research against cancer conducted in Lille, France in the SIRIC ONCO-Lille program. The fusion of the BioMEMS technology and Cancer research plans to take place in a hosting platform located on the Oscar Lambret Centre site, within the hospital-university campus, in order to be in close contact with medical teams. This platform provides an experimental environment in which IIS researchers can perform joint work in France. In other word, SMMiL-E is a “mirror” structure of LIMMS to welcome researchers from Japan and to organize French/Japanese teams performing tight joint research. The program also includes IEMN/CNRS and Lille-1 University. Currently, there are experimental/office space in IRCL (Institute of Cancer Research in Lille). Laboratory for SMMiL-E will be first installed in IRCL from 2016 and then moved to a new building built by 2020 with the support from the State, Local Government, CNRS, COL, Lille University and INSERM.

In April 2016, CIRMM was renewed under a new name “Centre for Interdisciplinary Research on Micro-Nano Methods”, and welcomed two new professors. The Centre named three axes of its activity, namely, “True-Nano Physics: exploitation of effects and phenomena based on nano to mesoscopic regime”, “Cyber Physical Systems Implementation”, and “Nano-Fabrication”. As the name implies, it puts emphasis on creating new methods, in terms of detection, processing, or implementation of époque making devices. The following schematic depicts the scope of our new centre of research. In April 2021, in addition, CIRMM was renewed as an internal center of IIS, while the main mission of CIRMM has been retained. In 2022, two new associate porfessors joined in CIRMM.



1-2. History of CIRMM

1999	CIRMM is proposed to Ministry of Education in Japan
	Preliminary discussion with CNRS/Science Pour l'Ingenieur(SPI) and positive response
Mar. 2000	Approval of the (Japanese/French) Diet
Apr. 2000	CIRMM is founded for 10-year term
Sep. 2000	CIRMM /CNRS in Paris opens in rue Capitaine Scott, LIP6/UPMC
Nov. 2000	Inauguration ceremony of CIRMM/CNRS Paris is held in CNRS/Headquarter in Paris
Oct. 2001	EPFL in Switzerland agrees to cooperate research activities with CIRMM
Mar. 2002	Twente University in the Netherlands agrees to cooperate research activities with CIRMM
Jul. 2002	Neuchatel University in Switzerland agrees to cooperate research activities with CIRMM
May 2003	KIMM in Korea agrees to cooperate research activities with CIRMM
Jul. 2003	University of Karlsruhe in Germany agrees to cooperate research activities with CIRMM
Aug. 2004	VTT Electronics and VTT Information Technology, Technical Research Center of Finland agreed to cooperate research activity with CIRMM
Apr. 2005	Seoul National University agrees to cooperate research activity with CIRMM
Nov. 2005	CIRMM contributes to establish NAMIS with CNRS, EPFL, IMTEK, SNU and VTT
Nov. 2006	National Tsing Hua University agrees to cooperate research activities with CIRMM
Jul. 2008	Initiation of Beans project founded by METI/NEDO Life-Beans Center and 3D-Beans Center are accommodated in IIS. Bio Nano Process collaboration Center was established.
Mar. 2009	CIRMM Evaluation Committee in Paris, France
Sep. 2009	CIRMM and VTT extended its Agreement of Joint Research
Apr. 2010	CIRMM is renewed for another 6 years / The official name became Center for International Research on Micronano Mechatronics
Oct. 2010	Academic Exchange Agreement signed with College of EECS in Seoul University
Dec. 2011	EU Project: EUJO-LIMMS (EUrope-Japan Opening of LIMMS) started for 4 years
Apr. 2012	Core-to-Core Program of the Japan Society for the Promotion of Science was launched by JSPS for 5 years
May 2012	EUJO-LIMMS Kick-off Meeting at IIS
Dec. 2012	JST Japanese-Taiwanese Cooperative Programme on 'Bioelectronics' started for 4 years with National Tsing Hua University: Neuron-on-CMOS-MEMS

	EUJO-LIMMS Info Day at EPFL
May 2013	LIMMS Workshop at Paris
Jun. 2013	Director General of Research & Technolgy Directorate at the European Commission, Mr. Robert Jan Smits, visited LIMMS
Sep. 2013	Workshop on Japan-Taiwan collaboration Research on Bio electronic at National TsingHuaUniversity in Hshinchu, Taiwan
Oct. 2013	EUJO LIMMS Info.Day at IMTEK in Freiburg, Germany
Dec. 2013	Workshop on Bio MEMS against Cancer at Centre Oscar Lambret in Lille, France
Jun. 2014	Inauguration Ceremony of SMMiL-E in Lille, France
	EUJO-LIMMS Workshop at VTT in Helsinki, Finland
Jul. 2014	Inauguration Symposium of CIBiS at IIS
Jan. 2015	20-year Aniversary of LIMMS at IIS
	Eujo-LIMMS Workshop at IIS
Apr. 2016	Foundation of new CIRMM (Centre for Interdisciplinary Research on Micro-Nano Methods)
Jun. 2016	Innovation show case at JST with the CNRS, LIMMS, CIBiS and the French embassy
Oct. 2016	Participation to Forum-X at the Ecole Polytechnique, Paris France
Dec. 2016	Joint Workshop of CIRMM/LIMMS/CIBiS on “International research leading to innovation and new technology bridging academics and societal demands”, at IIS
Feb. 2017	FEMO-ST Workshop “International Seminar LIMMS and FEMTO-ST and Partners”, Besancon France
Mar. 2019	LIMMS-Next PV Joint Energy Workshop at IIS
Nov. 2019	2019 NAMIS Marathon Workshop, Hsinchu, Taiwan
Nov. 2020	2020 NAMIS Marathon Workshop, Hybrid (Online and Hsinchu, Taiwan)
Apr. 2021	Renewal of CIRMM as Internal Center of IIS
Dec. 2021	2021 NAMIS Marathon Workshop, Hybrid (Online and Hsinchu, Taiwan)
Oct. 2022	NAMIS Core Members Online Meeting
Dec. 2022	2022 NAMIS Marathon Workshop, Hsinchu, Taiwan

1-3. History of NAMIS

Nov. 2005	CIRMM contributes to establish NAMIS with CNRS, EPFL, IMTEK, SNU and VTT
May 2006	The 1st NAMIS Workshop in IEF, Orsay and IEMN, Lille, France
Nov. 2006	The 2nd NAMIS Workshop in EPFL, Laudanne, Switzerland

Jun. 2007	The 3rd NAMIS Workshop in VTT, Oulu, Finland
Oct. 2007	International MEMS school for NAMIS students at IIS
Nov. 2007	The 4th NAMIS Workshop in Seoul, Korea
Apr. 2008	The 5th NAMIS Workshop in Freiburg, Germany
Sep. 2008	NAMIS International Autumn School 2008 at IIS, Tokyo The 6th NAMIS Workshop in Shonan Village, Japan University of Paris South and Tohoku University joined NAMIS as associated partners
Jun. 2009	The 7th NAMIS Workshop in Montreal, Canada.
Sep. 2009	NAMIS International Autumn School 2009 at Toulouse, France
Jun. 2010	The 7th NAMIS Workshop in Hsinchu Taiwan. University of Washington, Seattle, USA joined the network.
Oct. 2010	NAMIS International Autumn School 2010 at IIS, UTokyo, Japan
Jun. 2011	The 9th NAMIS Workshop in Paris, France
Sep. 2011	NAMIS International Autumn School 2011 at Neuchâtel – Arc-et-Senans - Besançon
May 2012	The 10th NAMIS Workshop at Sendai
Sep. 2012	NAMIS International Autumn School 2012 at IIS, UTokyo, Japan
Jul. 2013	The 11th NAMIS Workshop in Seattle
Sep. 2013	NAMIS International Autumn School 2013 in Seoul, Korea
Jun. 2014	The 12th NAMIS Workshop in Vietnum
Sep. 2014	NAMIS International Autumn School 2014 at National TsingHua University in Hshinchu, Taiwan
Jun. 2015	NAMIS International Autumn School 2015 in Montréal, Canada
Sep. 2015	The 13th NAMIS Workshop in Wroclaw University of Technology, Poland
Jul. 2016	The 14th NAMIS Workshop in University of Twente, The Netherlands
Sep. 2016	NAMIS International Autumn School 2016 in IIS, UTokyo, Japan
Jun. 2017	The 15th NAMIS Workshop in Daejeon, Korea
Oct. 2017	NAMIS International Autumn School 2017 in Freiburg, Germany
Jun. 2018	The 16th NAMIS Workshop in VTT-Oulu, Finland
Sep. 2018	NAMIS International Autumn School 2018 in University of Washington, Seattle, USA
Nov. 2019	2019 NAMIS Marathon Workshop, Hsinchu, Taiwan
Jan. 2020	Seoul National Univ.(SNU) / IIS, The Univ. of Tokyo Joint Workshop on Innovative Micro/Nano systems, Seoul, Korea

Nov. 2020	2020 NAMIS Marathon Workshop, Hybrid (Online and Hsinchu, Taiwan)
Dec. 2021	2021 NAMIS Marathon Workshop, Hybrid (Online and Hsinchu, Taiwan)
Oct. 2022	NAMIS Core Members Online Meeting
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1-4. Research Areas

- Micro/nano 3D fabrication technologies based on silicon process, precision, mechanical machining, printing, bonding, bio coating, soft lithography and self-assembly.
- Basic components such as micro actuators, micro fluidics, mirrors/gratings and nano structures.
- Fundamantals and application of various Microscopy
- Integration and packaging technologies; evaluation methods
- Applications especially in bio/medical systems, optics, wireless communication, IoT (internet of things) and nano technologies.
- Technology transfer to industry

1-5. Missions of Each Organization in CIRMM

- ❑ CIRMM for global networking activities in Micro- and Nano-systems
  - Stimulate exchanges by workshops, meetings, visits
  - Establish joint research projects between Japanese university network and foreign Universities/Laboratories.
- ❑ LIMMS (Laboratory for Integrated Micromechatronic Systems)
  - Accept CNRS related French researchers in IIS
  - Perform state-of-the art research on MEMS and applications.
  - EUJO-LIMMS (2011-2015): Extention to 4 European partners
  - SMMiL-E (2014-): Bio MEMS installation in Cancer Hospital in France
- ❑ NAMIS (International Research Group on Nano and Microsystems)
  - Stimulate research exchanges of twelve organizations:
    CNRS (France), EPFL (Switzerland), SNU (Korea), VTT (Finland), IMTEK (Germany), IIS (Japan), EPM (Canada), KIMM (Korea), UPE (France), NTHU (Taiwan), UW (USA), MESA+ (the Netherlands)
- ❑ IBEC (IIS/UTokyo Bureau for European Collaboration)

- Management of research projects in SMMIL-E and the support for IIS researchers visiting Lille.Technology transfer to clinical applications
- Networking with EU committees and with EU researchers
- Partial support of LIMMS administration in terms of CNRS related operation

1-6. CIRMM Offices and Oversea Laboratory

Tokyo Office

Address: 4-6-1 Komaba, Meguro-ku, Tokyo, 153-8505, Japan

The University of Tokyo, Institute of Industrial Science

Phone +81-3-5452-6248 at Prof. Fujita / Fax +81-3-3402-5078 (IIS main)

IIS/UTokyo Bureau for European Collaboration (IBEC)

Address: CNRS Délégation Nord Pas de Calais et Picardie, 2 rue des Canonniers CS60009, 59046 LILLE CEDEX, France

Phone: + 33 3 20 12 28 16

SMMiL-E Laboratory

Address: IRCL, Place de Verdun, 59000 Lille, France

(IRCL: Institut pour la Recherche sur le Cancer de Lille)

1-7. CIRMM Network

CIRMM’s micromechatronics (MEMS) research network in Europe, Asia, US/Canada and in Japan.

Centre de la National Recherche Scientifique (CNRS) France

http://www.cnrs.fr/

Ecole Polytechnique Federal de Lausanne (EPFL) Switzerland

http://www.epfl.ch/

University Paris Est (UPE) France

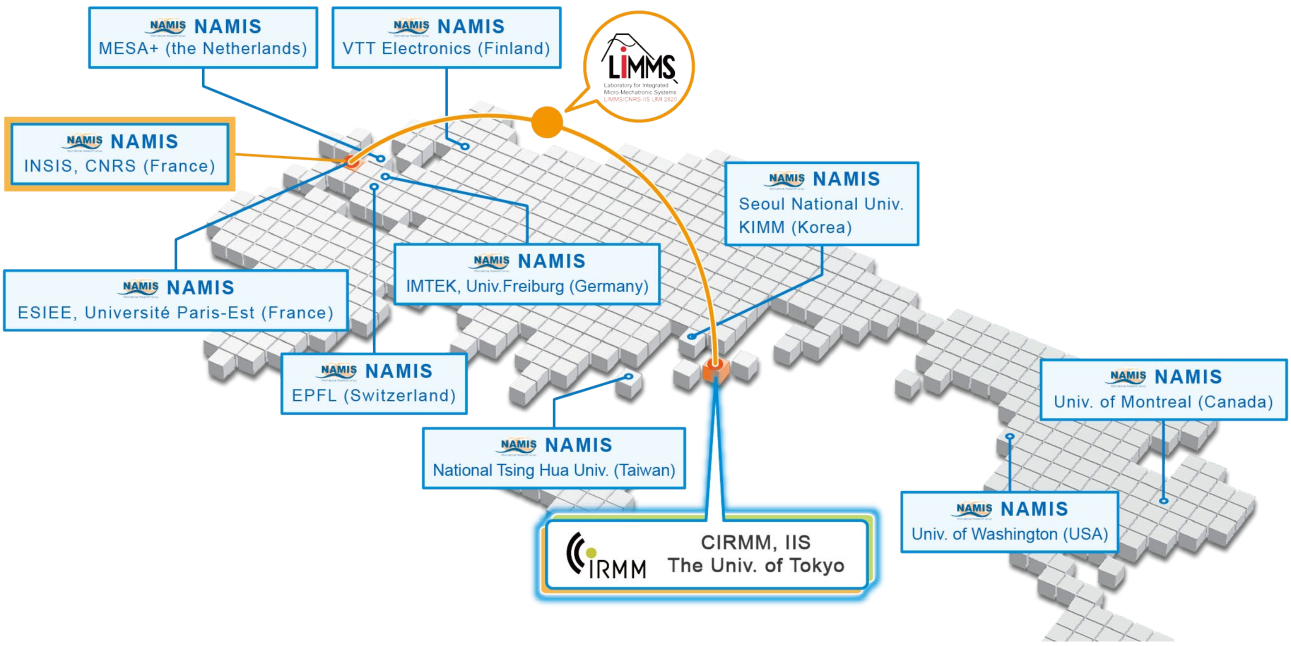
http://www.univ-paris-est.fr/fr/

VTT Technical Research Centre of Finland Ltd. Finland

http://www.hydromod.de/loleif/Participants/VTT/vtt.html

Department of Microsystem Engineering, University of Freiburg (IMTEK) Germany http://www.imtek.mml/material

Inter-University Semiconductor Research Center (ISRC)	South Korea
http://isrc.snu.ac.kr/	
Seoul National University (SNU)	South Korea
http://www.useoul.edu/	
Korea Institute of Machinery and Materials (KIMM)	South Korea
http://www.kimm.re.kr/main.php	
Polytechnique of Montreal	Canada
http://www.umontreal.ca/	
National Tsing Hua University (NTHU)	Republic of China (Taiwan)
http://www.nthu.edu.tw/english/index.php	
Tohoku University (TU)	Japan
http://www.tohoku.ac.jp	
University of Washington (UW)	USA
http://www.washington.edu/	
MESA+, Twente University	the Netherlands
http:// http://www.utwente.nl/mesaplus/	
Centre Oscar Lambret (COL)	France
http://www. centreoscarlambret.fr/	



CIRMM International Network

Other Related Organizations

Korea Institute of Industrial Technology — South Korea
http://english.itep.re.kr/

Research Center for Advanced Science and Technology, The University of Tokyo — Japan
http://www.rcast.u-tokyo.ac.jp/ja/

VLSI Design and Education Center (VDEC), The University of Tokyo — Japan
http://www.vdec.u-tokyo.ac.jp/English/index.html

Nanofabrication Platform, The Nanotechnology Platform Japan Program — Japan

1-8. Members

Name	Position	Research Field
TAKAHASHI, Takuji	Director and Professor	Nano-probing Technologies
TOSHIYOSHI, Hiroshi	Deputy Director and Professor	Optical MEMS & RF-MEMS
KAWAKATSU, Hideki	Professor	Coupling to the Nano Regime
KIM, Beomjoon	Professor	Micro Components & Systems
TAKAMIYA, Makoto	Professor	Integrated Power Management Circuits
MIZOGUCHI, Teruyasu	Professor	Understanding Role of Atom and Electron in Material
NOMURA, Masahiro	Professor	Nanoscale Heat Transfer and Thermoelectrics
TIXIER-MITA, Agnes	Associate Professor	Bio CMOS-MEMS Platforms
TOCHIGI, Eita	Associate Professor	Characterization of lattice defects
MATSUHISA, Naoji	Associate Professor	Interactive Electronic Devices

-Takahashi Laboratory Members:

SHIMADA, Yuuji (Technical Support Specialist)

YAMADA, Ayaka (Graduate Student)

LI, Shenwei (Graduate Student)

KOBAYASHI, Daichi (Graduate Student)

SATO, Jo (Graduate Student)

WEN, Sihan (Graduate Student)

YOGI, Naomi (Assistant)

-Toshiyoshi Laboratory Members:

ATAKA, Manabu (Research Associate)

TAKAHASHI, Takuya (Technical Support Specialist)

HASHIGUCHI, Gen (Research Fellow)

SASAKI, Naruo (Research Fellow)

ONO, Shimpei (Research Fellow)

YOKOKAWA, Ryuji (Research Fellow)

KAKUSHIMA, Kuniyuki (Research Fellow)

SEKIYA, Hidehiko (Research Fellow)

ISHIDA, Tadashi (Research Fellow)

KUMEMUEA, Momoko (Research Fellow)

YAMANE, Daisuke (Research Fellow)

NAGAI, Moeto (Research Fellow)

MITA, Makoto (Associate Research Fellow)

HIGO, Akio (Associate Research Fellow)

CHIU Wan-Ting (Associate Research Fellow)

YAMADA, Shunsuke (Associate Research Fellow)

HONMA, Hiroaki (Project Research Associate)

MENON, Vivek Anand (Project Research Associate)

IHIDA, Satoshi (Private Sector Collaborative Researcher)

ISAMOTO, Keiji (Private Sector Collaborative Researcher)

YI, Xiao (Private Sector Collaborative Researcher)

MITSUYA, Hiroyuki (Private Sector Collaborative Researcher)

ODA, Yutaro (Private Sector Collaborative Researcher)

SUGAHARA, Junpei (Graduate Student)

PUTRA, Refaldi Intro Dwi (Graduate Student)

AKAI, Yuto ((Graduate Student)

HU, Xingzhuo (Graduate Student)

FAURE, Gabriel (Graduate Student)
CHIKAMATSU, Kosuke (Graduate Student)
KOIZUMI, Hiroko (Assistant)

-Kawakatsu Laboratory Members:
KOBAYASHI, Dai (Research Associate)
SHIOZAKI, Yuriko (Assistant)
KIUCHI Kisa (Assistant)
OHTANI Mizuki(Assistant)
KATAYAMA Ryo (Graduate Student)
TOMOFUJI Kouji (Graduate Student)
ZHOU Zheyuan (Graduate Student)
ZHU Zhenyan (Graduate Student)
LI Yifan (Research Student)
LI Baitao(Research Student)

-Kim Laboratory Members:
PARK, JongHo (Assistant Professor)
SALLES, Vincent (Visiting Research Fellow)
HWANG, Gilgueng (Visiting Research Fellow)
TAKEUCHI, Kai (Private Sector Collaborative Researcher)
KOMA, Yosuke (Private Sector Collaborative Researcher)
TSURUMA,Yuko (Private Sector Collaborative Researcher)
BAO, LeiLei (Graduate student)
JING, Heyi (Graduate student)
SHOBAYASHI, Kotaro (Graduate student)
KUDO, Kota (Graduate student)
QIN, Boyu (Graduate student)
ZHANG, Jingzong (Graduate student)
DEBUN, Kotaro (Graduate student)
XU, Chensong (Graduate student)
XIE, Tingyu (Graduate student)
DELMAS, Zacharie((Internship Student)
SHOBAYASHI, Shujiro (Internship Student)
KINOSHITA, Rie (Technical assistant)
OKUDAIRA, Kazuko (Assistant)
KOIZUMI, Hiroko (Assistant)

-Takamiya Laboratory Members:

HATA, Katsuhiro (Research Associate)

SAI, Toru (Research Fellow)

INUMA, Toshiaki (Project Academic Specialist)

WANG, Ruizhi (Graduate Student)

ZHANG, Dibo (Graduate Student)

ZHANG, Haifeng (Graduate Student)

ZHOU, Haoxi (Graduate Student)

YANO, Hiroki (Graduate Student)

AOKI, Noel (Internship Student)

AMANO, Ayako (Assistant)

-Mizoguchi Laboratory Members:

SHIBATA, Kiyou (Assitant Professor)

FUKUDA, Atsushi (Technical staff)

XIE, Yaoshu(Postdoctral researcher)

KAWAGUCHI, Naoto (Graduate student)

HATA, Yuki (Graduate student)

CHEN, Poyen (Graduate student)

TAKAHARA, Izumi (Graduate student)

NISHIO, Kento (Graduate student)

LEE, Yebin (Graduate student)

-Nomura Laboratory Members:

ANUFRIEV, Roman (Project Associate Professor)

VOLZ, Sebastian (Visiting Research Fellow)

JALABERT, Laurent (Visiting Research Fellow)

ORDONEZ-MIRANDA, Jose (Visiting Research Fellow)

KOSEVICH, Yury (Visiting Research Fellow)

KIM, Byunggi (Project Research Associate)

YANAGISAWA, Ryoto (Project Research Associate)

WU, Yunhui (Project Researcher)

DIEGO, Michele (Project Researcher)

PIRRO, Matteo (Project Researcher)

HUANG, Xin (Graduate student)

KOIKE, Souta (Graduate student)

SHI, Hongyuan (Graduate student)

SINGH, Dhanishtha (Graduate student)

OGAWARA, Youhei (Graduate student)

WU, Xin (Visiting Associate Research Fellow)
YANG, Lei（Visiting Associate Research Fellow)
BIN, Chengwen (Visiting Associate Research Fellow)
CORAL, Maelie (Visiting Associate Research Fellow)
BARBIER–CHEBBAH, Felix (Visiting Associate Research Fellow)
TOYODA, Satoshi (Collaborative Researcher)
NAKAOKA, Toshihiro (Research Fellow)
XIONG, Shiyun (Research Fellow)
PARK, Keunhan (Research Fellow)
MAIRE, Jeremie (Research Fellow)
GUO, Yangyu (Associate Research Fellow)
ZHANG, Zhongwei (Associate Research Fellow)
TACHIKAWA, Saeko (Associate Research Fellow)
YAMASHITA, Yuichiro (Associate Research Fellow)
HARASHIMA, Junichi (Associate Research Fellow)
INADA, Yuta (Associate Research Fellow)
YOSHIDA, Yoshifumi (Associate Research Fellow)
OKOCHI, Erina (Assistant)

-Tixier-Mita Laboratory Members:
XU Tieying, (IIS Postdoctoral Fellow)
FAURE, Pierre-Marie (Visiting Associate Research Fellow)
MABIRE, Adrien (Internship Student)
KOIZUMI, Hiroko (Assistant)

-Tochigi Laboratory Members:
SATO, Takaaki (Project Researcher)
NAKAMURA, Toshiki (Graduate student)
IMAMIYA, Mana (Technical Assistant)
OGITA, Sakiko (Assistant)

-Matsuhisa Laboratory Members:
LIU, Yijun (PhD student)
SHIMURA, Tokihiko (Internship Student)
TOMINAGA, Taizo (Internship Student)
WANG, Liren (Internship Student)
MITOMO, Hinata (Graduate student)
ZHOU, Yuanyuan (Graduate student)
SEKI, Akiko (Assistant)

Affiliated members:

HIRAKAWA, Kazuhiko (Professor)

The University of Tokyo, Institute of Industrial Science
Research Area: Quantum Semiconductor Electronics

SAKAI, Yasuyuki (Professor)

The University of Tokyo, Institute of Industrial Science
Research Area: Organs and Biosystems Engineering

MITA, Yoshio (Associate Professor)

The University of Tokyo, School of Engineering
Research Area: Intelligent Electron Devices by MEMS Technology

SOMEYA, Takao (Professor)

The University of Tokyo, School of Engineering
Research Area: Flexible Electronics

SUGIYAMA, Masakazu (Professor)

The University of Tokyo, Research Center for Advanced Science and Technology
Research Area: Novel Micro-nano Fabrication Processes for Next-generation MEMS, Crystal Growth of III-V Semiconductors for High-efficiency Solar Cells

HIBARA, Akihide (Professor)

Tohoku University, Institute of Multidisciplinary Research for Advanced Materials
Research Area: Nano/Micro Chemical Measurements

HANE, Kazuhiro (Professor)

Tohoku University, Department of Nanomechanics
Research Area: Optical MEMS, especially Optical Micro-sensors and Integrated Optical Systems for Telecom

HASHIGUCHI, Gen (Professor)

Shizuoka University, Research Institute of Electronics
Research Area: Micromachining, Electret-based Energy Harvester

KONISHI, Satoshi (Professor)

Ritsumeikan University
Research Area: Microelectromechanical Systems (MEMS), Microrobotics, Biomedical Engineering

NOJI, Hiroyuki (Professor)

The University of Tokyo, School of Engineering
Research Area: Single Molecular Biophysics

Activity Highlight
April 2022- March 2023

April 13th, 2022
Light upon the mountain award

Prof. Naoji Matsuhisa won Light upon the mountain award for his contribution to the development of soft electronic devices for future healthcare sensors.

June 17th, 2022
Young Scientist Presentation Award from The Japan Society of Applied Physics

Prof. Naoji Matsuhisa won Young Scientist Presentation Award from The Japan Society of Applied Physics for his contribution in high-frequency and stretchable diodes.

June 29th, 2022
MIT Technology Review Innovators Under 35 Global

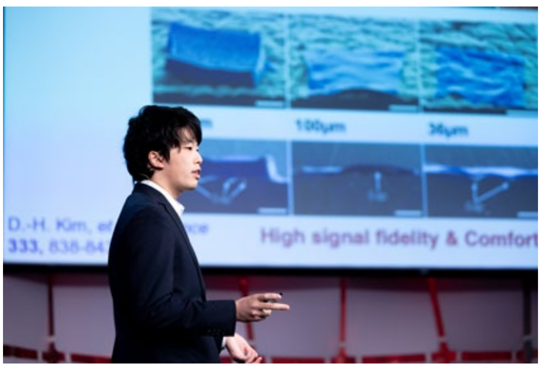
Prof. Naoji Matsuhisa was selected as one of the MIT Technology Review Innovators Under 35 Global for his contribution to high-frequency intrinsically stretchable diodes to realize future wearable devices. He was invited to give a seminar at EmTech MIT 2023, which is a flagship event by MIT Technology Review and held at MIT Media Lab.

Naoji Matsuhisa

Associate Professor

University of Tokyo; 2022 Innovator Under 35

Naoji Matsuhisa received his PhD degree from the Department of Electrical Engineering and Information Systems at the University of Tokyo in 2017. After his studies, he then worked as a postdoc at Nanyang Technological University in Singapore, and Stanford University in the U.S. In April 2020, he joined the Department of Electronics and Electrical Engineering at Keio University as an Assistant Professor. In April 2022, he became an Associate Professor at the Institute of Industrial Science at the University of Tokyo. He has focused on stretchable electronic materials and devices. His representative work includes a printable elastic conductor with a record conductivity and a high-frequency, intrinsically stretchable diode.



July 23rd, 2022
6th Phonon Engineering Workshop, Excellent Poster Award

Mr. Sota Koike won the 2020 Excellent Poster Award for his poster "Measurements of thermoelectric properties of SiGe membranes and improved performance of thermoelectric generators" presented at the 6th Phonon Engineering Workshop online (Jul. 22-23, 2022).

Oct. 5th, 2022
NAMIS Core Members Meeting

Although most activities within the framework of the NAMIS Network have been suspended since 2020 due to the COVID-19 pandemic, NAMIS Core Members Meeting was held online on October 5th to discuss plans for future activities. As a result, the NAMIS core members agreed to resume face-to-face NAMIS workshops from 2023.

November 22nd, 2022
Two parties of industrial collaborators are awarded the excellent paper awards.

At the IEEJ Sensor Symposium in Tokushima, the best paper award was given to the paper (CMOS image sensor) report in joint names with NHK Science & Technology Research Labs. Another excellent paper award was given to the paper (MEMS energy harvester) published in joint names with Saginomiya Seisakusho, Inc.


電気学会センサ・マイクロマシン部門　部門大会
第３９回「センサ・マイクロマシンと応用システム」シンポジウム
最優秀技術論文賞
SOI ウェハのハイブリッド接合を用いた
画素並列３層積層 CMOS イメージセンサ
*後藤　正英{1}, 本田　悠葵{1}, 難波　正和{1}, 井口　義則{1}, 更屋　拓哉{2},
小林　正治{2}, 日暮　栄治{3}, 年吉　洋{2}, 平本　俊郎{2}
{1}NHK 放送技術研究所, {2}東京大学, {3}東北大学
あなたは第３９回「センサ・マイクロマシンと応用システム」シンポジウムにおいて
厳正な審査の結果 最優秀技術論文賞に選ばれました
ここにあなたの研究がますます発展することを祈念して表彰いたします
2022 年 11 月 16 日
一般社団法人 電気学会
センサ・マイクロマシン部門
部門長　年吉　洋


電気学会センサ・マイクロマシン部門　部門大会
第３９回「センサ・マイクロマシンと応用システム」シンポジウム
優秀技術論文賞
カリウムイオンエレクトレット製デバイス量産化のための長期信頼性技術
*三屋　裕幸{1}, 芦澤　久幸{1}, 下村　典子{1}, 石黒　巧真{1}, 本間　浩章{2},
橋口　原{3}, 年吉　洋{2}
{1}鷺宮製作所, {2}東京大学, {3}静岡大学
あなたは第３９回「センサ・マイクロマシンと応用システム」シンポジウムにおいて
厳正な審査の結果 優秀技術論文賞に選ばれました
ここにあなたの研究がますます発展することを祈念して表彰いたします
2022 年 11 月 16 日
一般社団法人 電気学会
センサ・マイクロマシン部門
部門長　年吉　洋

December 4th-5th, 2022

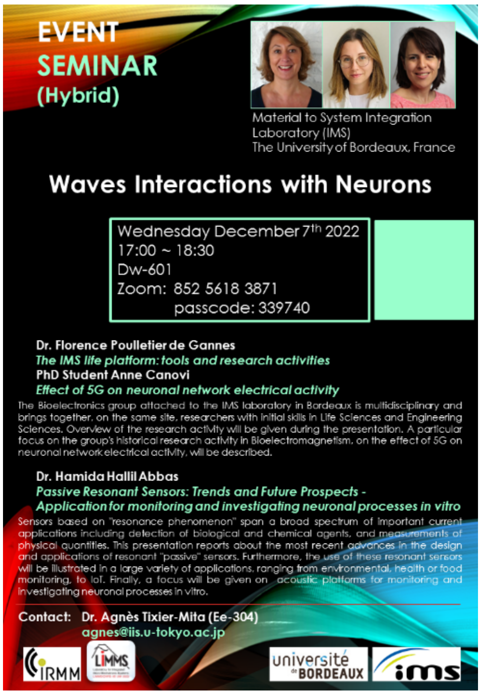
NAMIS Marathon Workshop was held in Taiwan from the 4th to the 5th of December 2022. More than ten researchers and students participated in the event from CIRMM and LIMMS. All the participants run the marathon and then gave aural presentations the following day. We are all very grateful to Professor Andrew Yeh, his staff and students for organizing such a wonder full event.

After the marathon

Closing with great success

December 7th, 2022
Hybrid seminar of the "Integration: from Material to Systems" laboratory of the University of Bordeaux.

A hybrid seminar on "Waves Interactions with Neurons" has been organized by Agnès Tixier-Mita, in the framework of a collaboration with the "**Integration: from Material to Systems" (IMS)** laboratory of the University of Bordeaux.



December 15th, 2022
PowerMEMS2022 Best Paper Award

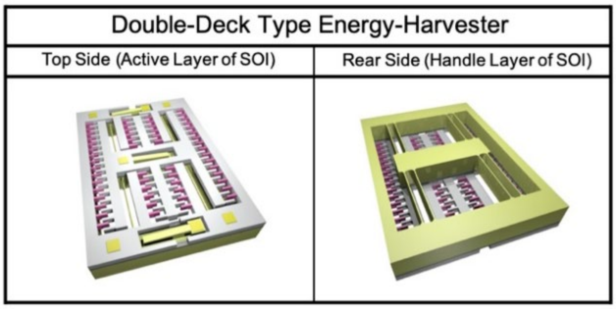
Dr. Ryoto Yanagisawa won the Best Paper Award for his paper "Planar-type nano-silicon thermoelectric generator over 100 μWcm^{-2}" presented at the 21st International Conference on Micro and Nanotechnology for Power Generation and Energy Conversion Application (PowerMEMS2022) in Salt Lake City, Utah, USA (Dec. 12 – 15, 2022). PowerMEMS conference is sponsored by Transducer Research Foundation and IEEE MEMS Technical Community and attracts broad audience.

February 3rd, 2023

Dr. Honma receives the Best Sensors & Materials Young Researcher Award 2022 for his work on MEMS energy harvester.

This award is presented to an author under the age of 40 who published the best paper of the year in the Journal Sensors & Materials.

Hiroaki Honma, Hiroyuki Mitsuya, Gen Hashiguchi, Hiroyuki Fujita, and Hiroshi Toshiyoshi, "Power Generation Demonstration of Electrostatic Vibrational Energy Harvester with Comb Electrodes and Suspensions Located in Upper and Lower Decks," Sensors and Materials, vol. 34, no. 4(3), 2022, pp. 1527-1538.




Best Sensors and Materials
Young Researcher Paper Award 2022

Awarded to

Hiroaki Honma

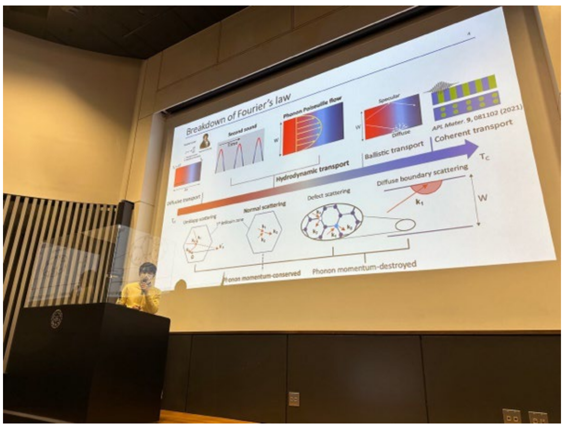
This is to certify that the paper entitled "Power Generation Demonstration of Electrostatic Vibrational Energy Harvester with Comb Electrodes and Suspensions Located in Upper and Lower Decks", written by Hiroaki Honma, Hiroyuki Mitsuya, Gen Hashiguchi, Hiroyuki Fujita, and Hiroshi Toshiyoshi, and published in Sensors and Materials, Vol. 34, No. 4(3), pp. 1527–1538, has been selected for the Best Sensors and Materials Young Researcher Paper Award 2022.

February 28, 2023

Makoto Ishida
Editor-in-Chief
Sensors and Materials, MYU K.K.

February 16th, 2023
CIRMM symposium for sharing research achievements of Doctoral/Master students

Doctoral and master students in CIRMM gave presentations about their research achievements for their thesis. In addition, there were presentations about three collaboration projects initiated by students in CIRMM. This symposium was initially scheduled in 2020, but was postponed because of COVID-19. More than 30 people, including professors and students, discussed various research conducted in CIRMM.



March 23rd, 2023
Telecommunications Systems Technology Award from the Telecommunications Advancement Foundation

Prof. Naoji Matsuhisa won Telecommunications Systems Technology Award from the Telecommunications Advancement Foundation for his paper “High-frequency and intrinsically stretchable polymer diodes” which was published in Nature.

Research Activities
April 2022 - March 2023

TAKAHASHI Laboratory

Current Research Activities 2022-2023

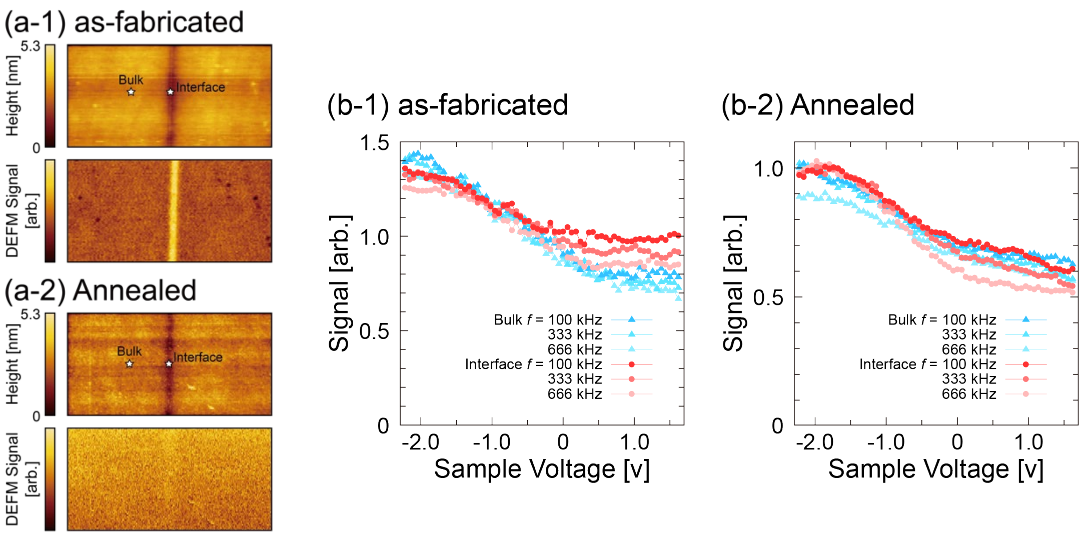
Takahashi Laboratory

1. Research Topics

1.1 Electrostatic Force Microscopy with Dual Bias Modulation Methods

Dual bias modulation electrostatic force microscopy (DEFM) enables us to examine depletion capacitance at high spatial resolution under various modulation frequencies. We used this DEFM to observe a cross section around an n-type Si/Si junction fabricated by surface-activated bonding, and the effect of thermal annealing on interfacial properties was investigated based on the DEFM signals corresponding to the $\partial C/\partial z$ signals.

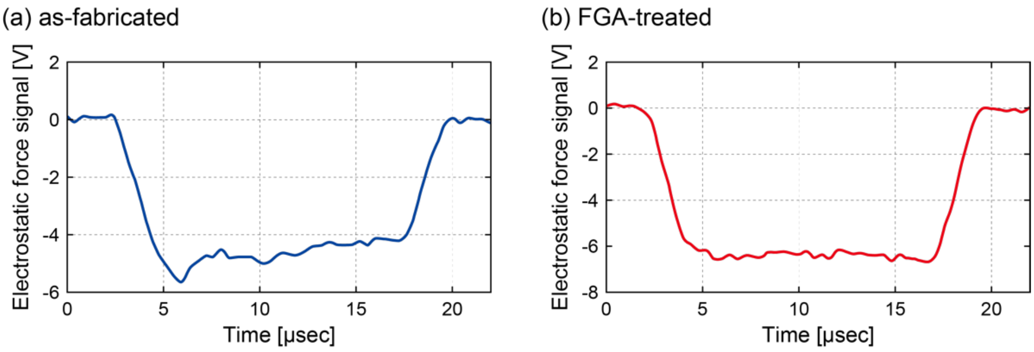
Figure shows topographic and DEFM signal images taken at null d.c. voltage together with DEFM signal spectra taken on the bulk and around the interface in the samples as fabricated and after annealing (5 min. at 1000 °C). From this figure, it was found that the DEFM signals were enhanced around the interface compared with those on the bulk in the as-fabricated sample, which was obvious especially under the positive d.c. sample voltages which correspond to the surface depletion condition for the n-type Si. Under the positive bias condition, in addition, clear dependence of the $\partial C/\partial z$ signal on the modulation frequency was observed at the interface, indicating the increase of capacitance under low modulation frequency condition. We attribute them to leaky electrical property of the interface due to highly dense defects originating from the fabrication process and to consequent enhancement of carrier transfer along the interface. On the annealed sample, to the contrary, the DEFM signals around the interface became very similar to those on the bulk. Therefore, we can conclude that the thermal annealing is very effective in reducing the defects at the interface.



1.2 Time-resolved Electrostatic Force Microscopy

Time-resolved measurements in electrostatic force microscopy (EFM) allows us to investigate charging and discharging properties of near-surface states in samples. By combining the intermittent bias application method which has been developed for improving a spatial resolution in electrostatic force detection in AFM and the pump-probe method which is widely used for time-resolved measurements, time-resolved EFM (Tr-EFM) has been constructed. In EFM measurements, however, the electrostatic force depends on an external d.c. bias as well as a contact potential difference (CPD) between a sample and tip. In order to nullify an influence of CPD, we also added a function of Kelvin probe force microscopy and realized real time control of base bias level in Tr-EFM measurements.

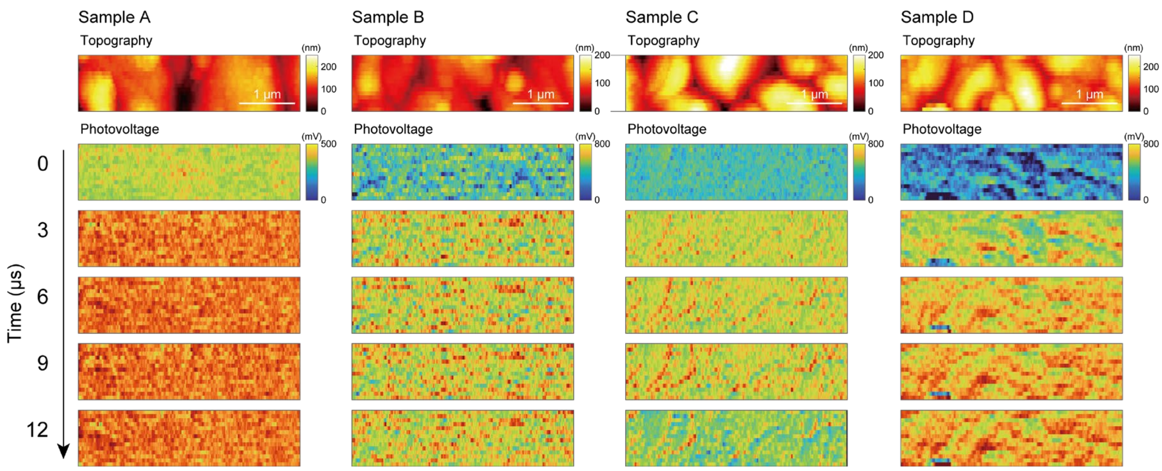
Figure shows waveforms of electrostatic force induced by a pulsed bias modulation taken on SiO_2/Si sample with and without FGA-gas treatment. As shown in the figure, electrostatic forces before and after the modulation were nearly null, indicating the base-bias-level control worked well. In addition, different responses were obtained: an overshoot was observed on the as-fabricated sample, being attributable to the response of charges accumulated at dense interface states between SiO_2 and Si.



1.3 Time-resolved Photo-assisted Kelvin Probe Microscopy on Solar Cells

Photo-assisted Kelvin probe force microscopy (P-KFM) allows us to investigate photovoltaic properties especially in solar cells, and if a temporal change in photovoltage can be observed by P-KFM, photocarrier dynamics will be analyzed in further detail. By replacing the electrical pump signal used in Tr-EFM above with an optical pump, time-resolved P-KFM has been realized.

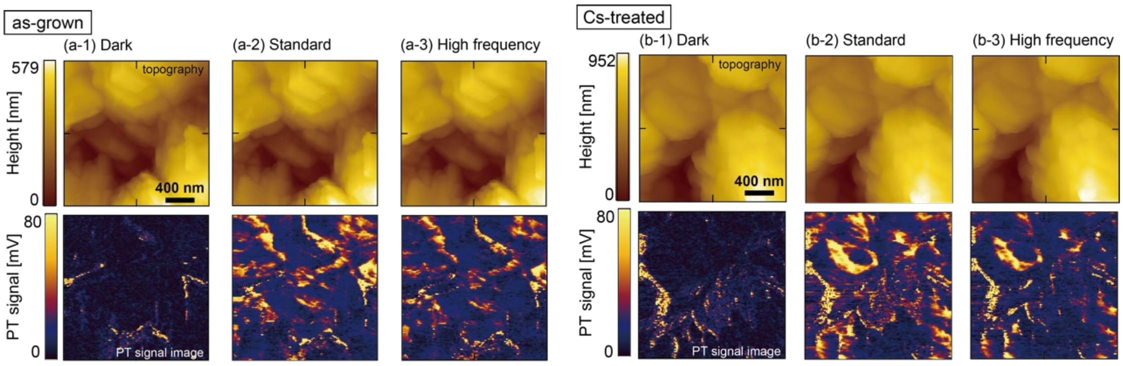
Figure shows temporal change of photovoltage just after the onset of the optical pump observed by P-KFM on Cu(In,Ga)Se_2 solar cells with different Ga contents, which enables us to investigate the difference in photovoltage rising process between grain interiors and their boundaries and also among various samples. Similarly, photovoltage decay process in these samples could be analyzed.



1.4 Photothermal Atomic Force Microscopy on Solar Cells

We performed the photothermal atomic force microscopy (PT-AFM), in which the periodical thermal expansion induced by the intermittent light illumination was detected by AFM, on the Cu(In,Ga)(S,Se)$_2$ [CIGSSe] materials to examine non-radiative recombination properties very locally.

In a multi-pulse (MP) modulation method in PT-AFM we originally developed, variable frequency measurements are realized. Especially under high frequency modulation, it is considered that heat sources near surface dominate the PT signal. The figure shows the PT signal images on as-grown CdS/CIGSSe samples and those after CsF-treatment taken under standard condition and under high frequency condition in MP method. From the comparison of them, we found the reduction of PT signal intensity on the Cs-treated sample under high frequency condition. Therefore, we concluded that the the CdS/CIGSSe interface was well passivated by the CsF treatment and consequently the non-radiative recombination was well suppressed there.



2. Research Achievements

2.1 Number of original journal papers: 5

2.2 International conference: 8

2.3 Domestic conference: 3

2.4 Number of patents: 0

3. List of awards

- none

4. Research Grants

4.1 Total number of research grants: 1

4.2 Number of collaboration research with industries: 1

4.3 List of major research grants (serving as Principal Investigator)

- Grant-in-Aid for Scientific Research “Dynamics of photo-generated carriers in multinary compound semiconductor solar cells investigated by photo-assisted nanoprobes” from JSPS

5. Education

5.1 Number of Ph.D. students (including current students): 1

5.2 Number of master students (including current students): 4

5.3 Number of other students: None

6. Publication list

Journal Papers

1. Tomoe Kuroiwa, Ryota Ishibashi, and Takuji Takahashi, "Time-resolved Photo-assisted Kelvin Probe Force Microscopy on $Cu(In,Ga)Se_2$ Solar Cells", *Japanese Journal of Applied Physics*, **61**, SL1004 (2022).
2. Ryota Fukuzawa, Jianbo Liang, Naoteru Shigekawa, and Takuji Takahashi, "Quantitative Capacitance Measurements in Frequency Modulation Electrostatic Force Microscopy", *Japanese Journal of Applied Physics*, **61**, SL1005 (2022).
3. Ayaka Yamada and Takuji Takahashi, "Effect of Cesium for $Cu(In,Ga)(S,Se)_2$ Solar Cells Using Photothermal Atomic Force Microscopy under Various Photoexcitation Conditions", *IEEE Journal of Photovoltaics*, **12** 1303-1307 (2022).
4. Kosuke Takiguchi, Le Duc Anh, Takahiro Chiba, Harunori Shiratani, Ryota Fukuzawa, Takuji Takahashi, and Masaaki Tanaka, "Giant Gate-controlled Odd-parity Magnetoresistance in One-dimensional Channels with a Magnetic Proximity Effect", *Nature Communications*, **13**, 6538 (2022).
5. Ryota Fukuzawa, Daichi Kobayashi, and Takuji Takahashi, "Accurate Electrostatic Force Measurements by Atomic Force Microscopy Using Proper Distance Control", *IEEE Transactions on Instrumentation and Measurement*, **72**, 1501408 (2023).

Conference Presentations

1. Ayaka Yamada and Takuji Takahashi, "Investigation of Cs Treatment Effect on $Cu(In,Ga)(S,Se)_2$ Solar Cell from Non-radiative Recombination Distribution under Various Photoexcitation Conditions Using Photothermal Atomic Force Microscopy", *the 49th IEEE Photovoltaic Specialists Conference*, Philadelphia, U.S.A., June (2022).
2. Takuji Takahashi, "Photo-assisted Scanning Probe Methods on Solar Cells", International Workshop on Micro- and Nano-Technologies for Energy, *Bio-engineering and Bio-sensing with JETMeE Workshop*, Toulouse, France, June (2022). [Invited]
3. Daichi Kobayashi, Naoteru Shigekawa, Jianbo Liang, and Takuji Takahashi, "Dual Bias Modulation Electrostatic Force Microscopy on n-type Si/Si Junctions Fabricated by Surface-activated Bonding", *The 22nd International Vacuum Congress*, Tue-C2-3, Sapporo, Japan, September (2022).
4. Ayaka Yamada and Takuji Takahashi, "Photothermal Atomic Force Microscopy on $Cu(In,Ga)(S,Se)_2$ Solar Cell Materials to Investigate CsF Treatment Effect", *The 22nd International Vacuum Congress*, Tue-C2-6, Sapporo, Japan, September (2022).
5. Takuji Takahashi, "Photo-assisted Scanning Probe Microscopy on Solar Cells", *LIMMS-IEMN Workshop*, Online, Dec. (2022). [Invited]
6. Ayaka Yamada and Takuji Takahashi, "Observation of Nonradiative Recombination on $Cu(In,Ga)(S,Se)_2$ Solar Cells using Photothermal Atomic Force Microscopy", *2022 NAMIS Marathon Workshop*, Hsinchu, Taiwan, Dec. (2022).
7. Daichi Kobayashi and Takuji Takahashi, "Annealing Effects on Si/Si Junctions fabricated by Surface-activated Bonding Investigated by Dual Bias Modulation Electrostatic Force Microscopy", *2022 NAMIS Marathon Workshop*, Hsinchu, Taiwan, Dec. (2022).
8. Jo Sato and Takuji Takahashi, "Verification of Time-resolved EFM Combined with KFM", *2022 NAMIS Marathon Workshop*, Hsinchu, Taiwan, Dec. (2022).

Japanese Journal of Applied Physics **61**, SL1004 (2022) **REGULAR PAPER**
https://doi.org/10.35848/1347-4065/ac5fbb

Time-resolved photo-assisted Kelvin probe force microscopy on $Cu(In,Ga)Se_2$ solar cells

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Photo-assisted Kelvin probe force microscopy (P-KFM) is very useful for examining photovoltaic characteristics especially on solar cells, although a time resolution in the conventional P-KFM was not sufficient for investigating carrier dynamics in time domain. In this study, we have introduced both an intermittent bias application method and a pump-probe method into P-KFM to realize time-resolved measurements at μsec order. Then the photo-carrier dynamics in $Cu(In,Ga)Se_2$ solar cells with different Ga compositions have been examined by our time-resolved P-KFM through direct observation of rising and decay waveforms of the photovoltage induced by the pulsed light illumination and through two-dimensional observation of photovoltage distributions at various time intervals between the optical pump and electrical probe pulses.

1. Introduction

Solar cells are very important for the decarbonized society and further improvement of the solar cell performance is desired. Among various materials for solar cells, $Cu(In,Ga)Se_2$ [CIGS] is suited especially for a thin film solar cell because of its very high optical absorption coefficient,[1)] and its bandgap can be varied between 1.04 and 1.68 eV, which covers an optimum bandgap (~1.4 eV) of a single junction solar cell,[2)] by changing the Ga composition.[3)] Up to now, a very high conversion efficiency up to 23.35% has been achieved in the CIGS solar cell.[4,5)] Since, however, typical CIGS used for the solar cell has a microcrystalline structure including small grains about 1 μm in size, microscopic investigation on the CIGS material is very helpful to understand photo-carrier dynamics in it.

For the microscopic investigation, scanning probe microscopy (SPM) like atomic force microscopy (AFM)[6,7)] is very suited because of its superior spatial resolution in topographic observation, and SPMs can also be applied to various electric characterization using a conductive AFM tip and some appropriate electric equipment. Especially by Kelvin probe force microscopy (KFM),[8,9)] where a surface potential can be determined through detection of an electrostatic force induced by external DC and AC voltages applied between a tip and a sample, the correlation between topography and electrical properties has been investigated on many kinds of materials and devices.[10–13)] When such SPMs are operated under light illumination, a photo-response of various samples can be investigated very locally, and also if such a method is applied to the solar cell material, dynamics of photo-generated carriers can be analyzed. Up to now, several attempts using photo-assisted Kelvin probe force microscopy (P-KFM)[14–29)] and photo-assisted conductive AFM[25,26,30)] have been reported to examine the open-circuit voltage and short-circuit current, respectively, which are two major properties of the solar cell. In addition, non-radiative recombination property of the photo-carriers has been investigated using a photothermal mode AFM (PT-AFM)[31–34)] and photothermal induced resonance technique in AFM.[35)]

In the SPM analyses, however, the intensity of photo-response signal is generally very weak, and consequently the signal should be amplified using an appropriate equipment like a current–voltage amplifier, lock-in amplifier and so on, while such an amplifier typically has narrow bandwidth (kHz order) and slow response time (msec order), being insufficient for direct investigation of the real-time dynamics of the photo-carriers. Thus there is a strong demand for development of a time-resolved measurement technique with a response time of much shorter than the msec order is strongly desired.

To realize the photovoltaic measurements in time domain by P-FKM, the method, in which the incident light is intermittently modulated at a certain frequency and the dependence of the temporally-averaged photo-response signal on the frequency is analyzed, has been proposed[19,21)] and demonstrated on a muli-crystalline Si solar cell,[19,21)] Si nanocrystal solar cell,[23)] organic solar sell,[24)] and CIGS solar cell.[29)] A similar modulation method was also applied to PT-AFM on the CIGS solar sell.[34)] In those methods, the time response was indirectly evaluated from the relationship between the temporally-averaged signal and the modulation frequency. If, to the contrary, the direct observation of the time response is realized, it will be much more efficient.

The group of the authors of the present paper has proposed the intermittent bias application method in KFM,[36,37)] where electrical pulses are applied between a tip and a sample only when the tip-end approaches a very vicinity of the sample surface within the oscillatory motion of the cantilever in the tapping mode, for the purpose of improving the spatial resolution in the potential measurement. By applying a similar pulse train as the probe together with another pulse train for excitation with a certain delay time and also by sweeping the delay time slowly, a pump-probe measurement has been realized.[38–40)] If light pulses are used for the photo-excitation in P-KFM, a time-resolved measurement of photovoltage will become possible.

In this study, we have applied such a pump-probe method in P-KFM to the CIGS solar cells with different Ga compositions because it is known that the actual solar cell characteristics depended on the Ga composition.[41)] We have examined the photo-carrier dynamics in those cells by the time-resolved P-KFM through direct observation of rising and decay waveforms of the photovoltage induced by the

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Quantitative capacitance measurements in frequency modulation electrostatic force microscopy

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We have proposed a method for quantitative capacitance measurements using frequency modulation electrostatic force microscopy (EFM) with a dual bias modulation method and demonstrated it on *n*- and *p*-type Si samples. First, we theoretically derived a conversion formula from a frequency shift of cantilever resonance in EFM into a capacitance value based on the parallel plate capacitor model, by which a pair of an EFM tip and a semiconductor sample is expected to be equivalently represented. Then the capacitance measurements were experimentally conducted on the *n*- and *p*-type Si substrates, and the acquired capacitance–voltage curves indicated that the obtained capacitance values were consistent with the expected ones and that the carrier densities evaluated from the depletion capacitances were also in good agreement with those evaluated by the conventional Hall effect measurements. From those results, the validity of our quantitative evaluation method has been well confirmed.

1. Introduction

Scanning probe microscopy (SPM) is an excellent analytical tool for sample property at high spatial resolution. In Atomic force microscopy (AFM),[1] many kinds of force acting between a tip and sample, such as van der Waals, chemical bonding, electrostatic, and magnetic forces, can be detected, and AFM has also been utilized to examine electronic materials and devices. In scanning capacitance microscopy (SCM)[2] and scanning nonlinear dielectric microscopy (SNDM)[3] that often use a capacitance sensor, the capacitance between the tip and sample is indirectly investigated, but the actual capacitance value itself is hard to evaluate because the tip-sample capacitance, whose value is considered to be aF (10^{-18} F) order,[4,5] is often hidden by a stray capacitance around it. In spite of such a problem, a capacitance change induced by an external bias voltage can be detected in SCM and SNDM as a $\partial C/\partial V$ signal,[6–11] where C and V are capacitance and voltage between the AFM tip and the sample, respectively, and a carrier density in a semiconductor can be evaluated quantitatively from the $\partial C/\partial V$ signal by comparison with that acquired on an appropriate reference sample with a known carrier density. In scanning microwave impedance microscopy (SMIM),[12–16] on the other hand, the capacitance value can be quantitatively evaluated through impedance analysis by calibrating it also using an appropriate reference sample, but the applicable frequency is only around GHz because of usage of microwave. Hence, broadband and variable frequency measurements are difficult in SMIM, although some slow behaviors of electric charges, such as response of isolated charge traps in a semiconductor and charge migration in an organic material, are very interesting subjects to investigate locally. Using electrostatic force microscopy (EFM),[17] such broadband and variable frequency measurements of the capacitance-related signals have been realized by multiple bias modulation methods[18–21] or amplitude modulated voltage application techniques,[22,23] and an influence of surface states has been investigated.[20,24] Especially when the electrostaticforce modulated by the external bias between the tip and sample is detected by the frequency shift of the cantilever resonance,[25,26] which is a similar way to frequency modulation (FM)-AFM[27] and is referred to as FM-EFM here, the detected force is mainly dominated by the very local force acting on the tip itself by reducing a contribution from background force acting on the body of cantilever,[28] from which the tip-sample capacitance can be sensitively evaluated. When, however, we want to quantitatively evaluate a capacitance on a semiconductor sample by FM-EFM, the following point should be carefully taken into account:

(i) The capacitance depends on the applied bias and has to be treated as a function of V instead of a constant.
(ii) Since the observed frequency shift represents the force gradient, $\partial F/\partial z$, an appropriate conversion formula from the frequency shift into the capacitance is required.
(iii) An actual shape of the tip end is unknown.

Here, F and z are an induced force and distance between the tip and sample, respectively. As for (i), S. Hudlet et al. derived the exact formula for a metallic tip and a semiconductor in an one-dimensional model,[29] by considering charges distributed inside the semiconductor together with those accumulated at the surface of the tip, while (ii) and (iii) have been still open questions.

This paper aims at finding a formula to convert the frequency shift value into the capacitance on the semiconductor with taking the frequency response of capacitance into account, which can be applied to (ii). Using the formula we propose based on a parallel plate capacitor model, the tip-sample capacitances have been evaluated from the frequency shift values obtained by FM-EFM on *n*- and *p*-type Si substrates, and, from the acquired capacitance values, carrier densities in those substrates have been calculated in order to verify our model and formula.

2. Formulation of frequency shift induced by electrostatic force between metallic tip and semiconductor

First, we suppose a parallel plate capacitor, as illustrated in Fig. 1, to replace the actual tip-sample system. Since the frequency shift measured in FM-EFM is sensitive to the force gradient around the tip end[28] as described above, a metallic

Effect of Cesium for $Cu(In,Ga)(S,Se)_2$ Solar Cells Using Photothermal Atomic Force Microscopy Under Various Photoexcitation Conditions

Ayaka Yamada and Takuji Takahashi

***Abstract*—We have performed photothermal (PT) measurements by atomic force microscopy, by which nonradiative recombination of photoexcited carriers can be examined very locally, on $Cu(In,Ga)(S,Se)_2$ (CIGSSe) materials in order to investigate CsF-treatment effects. From the PT signal images taken under various photoexcitation conditions and by comparing those images taken on CIGSSe samples with or without the CsF-treatment, we have found the possibility that the nonradiative recombination centers, which should exist along grain boundaries and may be distributed at a CdS/CIGSSe interface, are well passivated by the CsF-treatment.**

***Index Terms*—Atomic force microscopy (AFM), CIGS, CsF-treatment, nonradiative recombination, phtothermal.**

I. Introduction

SOLAR power generation is one of the promising renewable energy sources which are essential to realize a sustainable society, and solar cells play a very important role for power conversion from solar energy. A $Cu(In,Ga)(S,Se)_2$ (CIGSSe) solar cell, typically used as a thin film solar cell, exhibits excellent performance, such as long-term stability [1] and strong irradiation resistance [2], compared with other solar cells. In 2013, Chirilă et al. [3] reported the achievement of an energy conversion efficiency of 20.4% on the CIGSSe solar cell by alkali fluoride (NaF and KF) postdeposition treatment (PDT), indicating incorporation of alkali metals into the CIGSSe thin film improved the performance of the CIGSSe solar cells very well. Afterwards, the CIGSSe solar cells with the conversion efficiency over 20% have been realized using PDT by alkali fluorid, such as KF, RbF, and CsF [4], [5], [6], [7], [8], [9], and the conversion efficiency up to 23.35% has been achieved on the CIGSSe solar cell by means of the CsF-treatment [10].

In recent years, many studies to investigate the mechanism of alkali-metal PDT have been conducted for further improvement of the CIGSSe solar cell performance [11], [12], [13], [14], [15], [16], [17], [18], [19], [20], and it was found that the incorporation of Cs contributed to the development of high-efficiency CIGSSe solar cells more effectively than the incorporation of Na and K, while the mechanism behind the efficiency-boosting by CsF has not been fully understood [21].

Similarly to the case of other alkali metals, the presence of Cs at grain boundaries (GBs) has been revealed [17], [18], [22], [23]. On the other hand, it is still required to investigate the effect of Cs on an interface between an absorber and buffer, while it was found that other alkali metals formed alkali-In-Se_2 ($AlkInSe_2$) compounds at the interface [24], [25].

In this study, the CsF-treatment effects on the CIGSSe material covered with a CdS thin film have been investigated by photothermal atomic force microscopy (PT-AFM) [26], [27], [28], [29], [30], by which nonradiative recombination property induced by a photoexcitation can be investigated very locally, under various photoexcitation conditions. In particular, the CsF-treatment effect on the GBs in CIGSSe and on the CdS/CIGSSe interface has been examined in terms of the nonradiative recombination.

II. Experimental

Fig. 1(a) shows our experimental setup for PT-AFM based on a commercial atomic force microscopy (AFM) system (SPA-300HV/SPI4000; Hitachi High-Tech Science Corporation, Japan) operated in the intermittent contact mode to observe topography in high vacuum ($\sim 10^{-5}$ Pa) at room temperature. A piezo-resistive cantilever whose nominal spring constant, fundamental resonant frequency, and quality factor in vacuum were around 40 Nm^{-1}, 400 kHz, and 3000, respectively, was used, instead of the optical deflection method which is widely used in AFM, to avoid stray light illumination onto a sample surface.

In PT-AFM, a sample surface is illuminated by an intermittently modulated light. If the sample absorbs the incident light, electrons and holes are generated, and also if those carriers recombine nonradiatively, heat is generated and thermal expansion occurs in the sample. When the light is turned off, the heat begins to dissipate and the sample begins to shrink. Therefore, under the intermittent light illumination, the thermal expansion under light illumination and the shrinkage in darkness occur periodically,

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Giant gate-controlled odd-parity magnetoresistance in one-dimensional channels with a magnetic proximity effect

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According to Onsager's principle, electrical resistance R of general conductors behaves as an even function of external magnetic field $\mathbf{B}$. Only in special circumstances, which involve time reversal symmetry (TRS) broken by ferromagnetism, the odd component of R against $\mathbf{B}$ is observed. This unusual phenomenon, called odd-parity magnetoresistance (OMR), was hitherto subtle (< 2%) and hard to control by external means. Here, we report a giant OMR as large as 27% in edge transport channels of an InAs quantum well, which is magnetized by a proximity effect from an underlying ferromagnetic semiconductor (Ga,Fe)Sb layer. Combining experimental results and theoretical analysis using the linearized Boltzmann's equation, we found that simultaneous breaking of both the TRS by the magnetic proximity effect (MPE) and spatial inversion symmetry (SIS) in the one-dimensional (1D) InAs edge channels is the origin of this giant OMR. We also demonstrated the ability to turn on and off the OMR using electrical gating of either TRS or SIS in the edge channels. These findings provide a deep insight into the 1D semiconducting system with a strong magnetic coupling.

Investigation of new magnetoresistance (MR) phenomena is an important issue in condensed matter physics, magnetism, and spintronics. For example, the discovery of giant MR[1,2] and tunneling MR[3,4] paved the way to the creation of non-volatile storage and memory devices. Generally, these MRs are even functions of external magnetic field B according to Onsager's principle[5]. However, it may not be the case when time reversal symmetry (TRS) is broken by magnetism in the system. The odd-parity MR (OMR) in a linear-response regime has been observed in systems where TRS is violated[6–9]. (See also Supplementary Table 1). To explain these OMR phenomena, various possible origins were proposed, including non-trivial Berry curvature, magnetic moments, side jump mechanism[10], and coexistence of spin–orbit interaction (SOI) and ferromagnetic coupling in a helical magnet[11]. Even in such rare systems, the OMR magnitude is typically very subtle (the magnitude reported thus far is at most 2%). In addition, these systems reported thus far are metallic, which hinders the control of OMR by external means such as electrical gate voltage.

In this Article, we report a giant and gate-controlled OMR in the edge transport channels of an InAs thin film interfaced with a ferromagnetic semiconductor (FMS) (Ga,Fe)Sb[12–14] layer (see Fig. 1a). The OMR is found to be unprecedently large; the resistance change is 27% of the total resistance when the $\mathbf{B}$ direction is reversed between ±10 T at $I = 1\ \mu A$. This is striking, considering that the SOI of InAs is much smaller than other materials such as $SmCo_5$ and pyrochlores in which OMR was

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Accurate Electrostatic Force Measurements by Atomic Force Microscopy Using Proper Distance Control

Ryota Fukuzawa, Daichi Kobayashi, and Takuji Takahashi

***Abstract*— Electrostatic force microscopy (EFM) enables us to examine the local electrostatic force between an atomic force microscopy (AFM) tip and sample surface, from which an electrical capacitance between them and surface potential can be evaluated, by applying direct current (dc) and alternating current (ac) voltages. From the dependences of the electrostatic force on the dc voltage and the frequency of ac voltage in EFM, carrier density, carrier type, and deep-level states in a semiconductor can also be investigated. Since EFM is basically operated as an AFM, special care should be taken in an effect of the electrostatic force on a distance control between the tip and the sample, and robust distance control is necessary even under the strong electrostatic force to realize proper measurements of both topography and electrostatic force. In this article, we have examined the effectiveness of the usage of the oscillation amplitude of a cantilever as a feedback target for the distance control, which is referred to as ΔA-mode operation, while the cantilever oscillation frequency is always kept at its primary resonance, like the conventional frequency modulation AFM (FM-AFM) which uses a resonant frequency shift as the feedback target. First, we have verified that a distance change due to the strong electrostatic force was significantly reduced compared with FM-AFM. Second, we have confirmed that the ΔA-mode operation in EFM realized proper measurements of the dependence of a tip–sample capacitance including a surface depletion capacitance on the dc voltage and the frequency of ac voltage, indicating good robustness of the method against the strong electrostatic force.**

***Index Terms*— Atomic force microscopy (AFM), electrostatic force, semiconductor.**

I. Introduction

ATOMIC force microscopy (AFM) [1] was developed for the investigation of surface structure for various materials with atomic scale [2], [3], and there are a lot of applications of AFM for electrical investigation with high spatial resolution [4]. In the electrostatic force microscopy (EFM) [5] and Kelvin probe force microscopy (KFM) [6], where an electrostatic force induced between an AFM tip and the sample surface is measured, an electrical capacitance and contact potential difference between them can be investigated, respectively. The scanning capacitance force microscopy (SCFM) [7], which is another application of EFM, can be applied to dopant profiling through $\partial C/\partial V$ measurements, where C and V are the capacitance and the voltage between the AFM tip and the sample, respectively. For accurate measurements by EFM, KFM, and SCFM, a tip–sample distance has to be appropriately regulated because the electrostatic force and capacitance surely depend on it.

In frequency modulation AFM (FM-AFM) [8], an interactive force between the tip and the sample can be evaluated from a shift of the resonant frequency of the cantilever detected by a frequency demodulator with high sensitivity especially in high-Q environment, where Q is a quality factor. This FM technique can be applied to the electrostatic force detection in EFM [9], [10], and it is referred to as FM-EFM in this article. A bandwidth in a typical frequency demodulator is designed to be very narrow (~kHz) to ensure a sufficient sensitivity [11] in FM-AFM, but it limits the frequency bandwidth in FM-EFM measurements. The narrow bandwidth makes deep-level investigation by FM-EFM difficult because it requires variable frequency measurements of capacitance. To overcome such a problem, we have developed the dual-bias modulation EFM (DEFM) [12], [13], [14], which is an application of high-frequency EFM [15], [16] (HF-EFM), using detection of high-order frequency component of the electrostatic force. In addition, amplitude-modulated voltage application [17], [18], [19] is applicable to variable frequency measurements. In these EFM techniques, electronic properties, including carrier density and deep-level states, can be investigated from the dependences of the electrostatic force on direct current (dc) voltage and the frequency of alternating current (ac) voltage, and it is necessary to keep the tip–sample distance constant under various conditions. In FM-AFM, however, the oscillation frequency of a cantilever is kept at its resonant frequency by the oscillation loop system [8], and the tip–sample distance is controlled to make the resonant frequency shift, Δf, constant, while the frequency shift is caused not only by the van der Waals force but also by the electrostatic force. If an electrostatic force image is simultaneously taken with a topographic image in the single scan mode, the former

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TOSHIYOSHI Laboratory

Current Research Activities 2022-2023

Toshiyoshi Laboratory

1. Research Topics

1.1 Double-Deck MEMS Electrostatic Vibrational Energy Harvester

Lowering internal impedance, i.e., stray capacitance and resistance, is essential to enhance the output power of microelectromechanical system (MEMS) vibrational energy harvesters particularly when the structure is made of a silicon-on-insulator (SOI) wafer which has a distributed stray capacitance across the buried oxide (BOX) layer. Instead of using silicon as a material for electrical interconnection, we used an out-of-plane aluminum bonding wire to complete the on-chip electrical interconnection between the contact pads on the chip frame and the suspended movable electrodes. Compared with the conventional design using silicon-based interconnections, the maximum deliverable power has been enhanced almost six-fold because of the reduced stray capacitance between the SOI layer and the substrate, as well as reduced internal electrical resistance. The mechanical stability of the bonded wire during the excitation vibration has also been experimentally confirmed. A typical power density of 763 μW/cm^3 is obtained when excited by 0.5 G (1 G = 9.8 m/s^2) sinusoidal vibrations at 157 Hz.

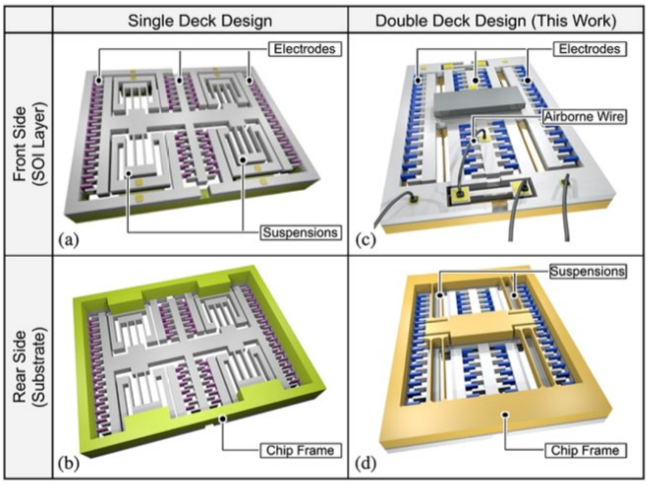

Figure 1. Double-deck MEMS vibrational energy harvester [J6].

1.2 Normally Off Time-of-event logging system using battery-less sensor

A time-of-event logging system has been demonstrated to detect the status changes of monitored objects. The event-driven circuit has a current capacity of as large as 33 mA to drive the load, which is sufficient to transmit data by using a wireless sensor node. A PVDF film as an event-driven switch is used to turn on the circuit by application of the mechanical impact on it. The energy consumption is 568 μJ, which is sufficiently small to log the time data every 10 s with an energy harvester of 10-μA current capacity.

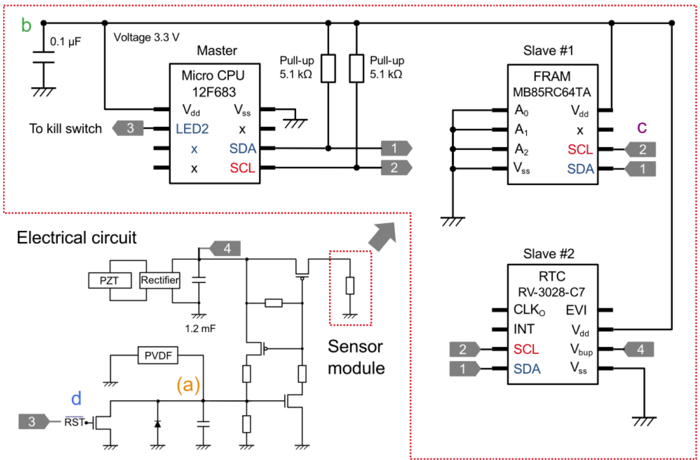

Figure 2. Electrical circuit of the time logging system using battery-less sensor [J1]

1.3 MEMS Switching Voltage Regulator using Electret Relay

We report on a DC/DC step-down switching regulator using a MEMS switch for the first time. The output voltage developed in a storage capacitor is used as a feedback control voltage for the electrostatic actuator that drives a micromechanical contact switch inserted between the rectifier and the capacitor. The switch is configured to be normally-on owing to the electrostatic potential remaining in the electret film on the electrodes. When the output voltage exceeds and the feedback control voltage cancels the built-in potential of the electret, the differential voltage acting on the actuator becomes small and the contact is opened, thereby decoupling the rectifier from the storage capacitor. When the output voltage becomes low, on the other hand, the net differential voltage becomes large and the contact is closed again, thereby reconnecting the rectifier to the capacitor and recovering the voltage. Fast repetition of this sequence maintains the DC voltage of the storage capacitor at a constant level. For demonstration, an electret film that is pre-charged to 240 V is used to regulate the output voltage in a range between 12 V and 14 V. Different from the switching regulators using transistors, the MEMS regulator does not require any bias current for operation, being suitable as a low-power voltage regulator for environmental energy harvesters

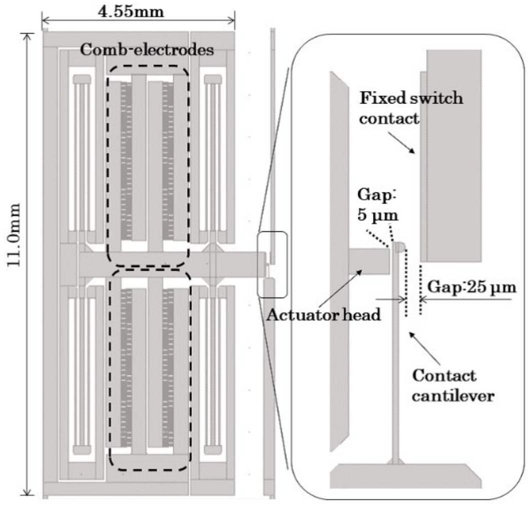

Figure 3. Structure of MEMS switching voltage regulator using a normally-on electret relay [J8].

1.4 Effect of Hydrogen on Potassium-ion Electrets

Potassium-ion electrets, which are mainly composed of amorphous silica and potassium atoms, can accumulate negative charges and are expected to be useful for future vibration powered generators. According to previous research, the origin of the accumulation of negative charges was revealed to be related to the presence of SiO_5 structures in amorphous silica. In general, Si–O bonds are so stable that potassium-ion electrets are expected to retain a high electric potential longer than previously studied materials and are therefore being studied intensively for practical use. However, charge decay phenomena were observed in previous experiments, and this presents serious problems. In this study, we focus on the role of hydrogen which is considered to be one of the origins of charge decay and have investigated the behavior of hydrogen in potassium-ion electrets and the mechanism of the charge decay using first-principles calculations. As a result, we found that hydrogen atoms can create a positively charged characteristic local structure or break SiO_5 structures and therefore cause degradation of the negative charge storing ability of potassium-ion electrets.

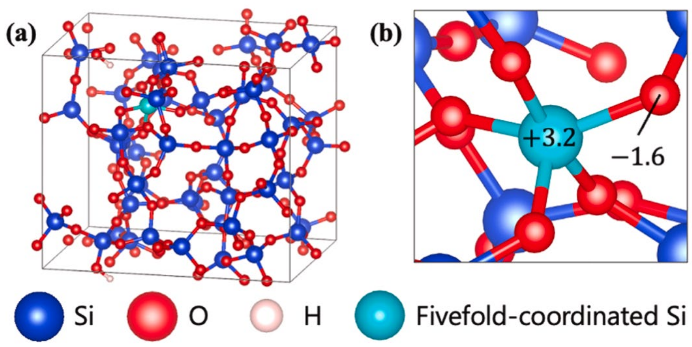

Figure 4. Characteristic fivefold-coordinated silicon structure as a source of negatively charged electret after displacing the potassium atom (molecular dynamics calculation) [J2].

2. Research Achievements (April 2022 – March 2023)

2.1 Number of original journal papers: 7

2.2 International conference: 14 (including 4 invited presentation)

2.3 Domestic conference: 2 (including 5 invited presentations)

2.4 Number of patents: 3

3. List of awards

1. H. Honma, Best Sensors&Materials Young Researcher Paper Award, 2023-02-28
2. H. Mitsuya et al., Excellent Paper Award, IEEJ Sensor Symposium, 2022-11-16
3. H. Goto et al., Best Paper Award, IEEJ Sensor Symposium, 2022-11-16

4. Research Grants

4.1 Total number of research grants: 4

4.2 Number of collaboration research with industries: 4

4.3 List of major research grants (serving as Principal Investigator)

- JST CREST Number JPMJCR19Q2, "Perpetual Electronics"
- JSPS Grant in Aid 21H01271, "Electret Degradation Mechanism"
- MIC 0155-0189, "Miniaturization of Atomic Clock"

5. Education

5.1 Number of Ph.D. students (including current students): 0

5.2 Number of master students (including current students): 6

6. Publication list

Journal Paper (selected, *attached)

J1. * Shunsuke Yamada and Hiroshi Toshiyoshi, "A Normally off Time-of-Event Logging System Triggered by A Battery-less Sensor," Sensors and Actuators: A. Physical, vol. 354, 2023, p.114306.

J2. * Yoshiki Ohata, Masaaki Araidai, Takuma Ishiguro, Hiroyuki Mitsuya, Hiroshi Toshiyoshi, Yasushi Shibata, Gen Hashiguchi, and Kenji Shiraishi, "Effect of hydrogen atoms on potassium-ion electrets used in vibration-powered generators," Materials Science in Semiconductor Processing, vol. 157, April 2023 (special issue: Control of Semiconductor Interfaces (ISCSI-IX)).

J3. * Yoshiki Ohata, Toru Nakanishi, Kenta Chokawa, Masaaki Araidai, Takuma Ishiguro, Hiroyuki Mitsuya, Hiroshi Toshiyoshi, Yasushi Shibata, Gen Hashiguchi, and Kenji Shiraishi, "Improvement of the reliability of potassium-ion electrets thorough an additional oxidation process," Applied Physics Letters, vol. 121, 2022, pp. 243903-1~5.

J4. Makoto Mita and Hiroshi Toshiyoshi, "A 2-Dimensional MEMS Optical Scanner for Landing Laser Rader," Koku-Uchu-Gijyutsu (Aerospace Technology Japan, the Japan Society for Aeronautical and Space Sciences)、vol. 21, 2022, pp. 62-67. (in Japanese)

J5. Dongchen Zhu, Anne-Claire Eiler, Satoshi Ihida, Yasuyuki Sakai, Hiroshi Toshiyoshi, Agnès Tixier-Mita, and Kikuo Komori, "Real-time Measurement of Pancreatic ß Cell Electrophysiology with Fluorescent Bioimaging Based on High-resolution Thin-film Transistor Microelectrode Arrays," IEEJ Trans. SM, vol. 142, no. 10, 2022, pp. 266-272.

J6. * Hiroaki Honma and Hiroshi Toshiyoshi, "Double-Deck MEMS Electrostatic Vibrational Energy Harvester with Airborne Interconnection," IEEJ Trans. SM, vol. 142, no. 9, 2022, pp. 215-219.

J7. Hiroyuki Mitsuya, Katsufumi Hashimoto, Kai-Chun Chang, Hisayuki Ashizawa, Noriko Shimomura, Tatsuki Momma, Hiroaki Honma, Gen Hashiguchi, Hiroshi Toshiyoshi, and Tomoki Shiotani, "Low-Power Frequency Monitoring System for Bridge using MEMS Vibrational-Energy

Harvesting Sensor," IEEJ Transaction on Sensors and Micromachines, vol. 142, no. 7, 2022, pp. (in Japanese)

J8. * Mizuki Morikawa, Yasushi Shibata, Hiroshi Toshiyoshi, and Gen Hashiguchi, "MEMS switching voltage regulator using a normally-on electret relay," IEEE/ASME Journal of Microelectromechanical Systems (JMEMS), vol. 31, no. 3, 2022, pp. 424 - 434.

J9. Hiroaki Honma, Hiroyuki Mitsuya, Gen Hashiguchi, Hiroyuki Fujita, and Hiroshi Toshiyoshi, "Power Generation Demonstration of Electrostatic Vibrational Energy Harvester with Comb Electrodes and Suspensions Located in Upper and Lower Decks," Sensors and Materials, vol. 34, no. 4(3), 2022, pp. 1527-1538.

J10. Hiroaki Honma, Yukiya Tohyama, Hiroyuki Mitsuya, Gen Hashiguchi, Hiroyuki Fujita, and Hiroshi Toshiyoshi, "Power Enhancement of MEMS Vibrational Electrostatic Energy Harvester by Stray Capacitance Reduction," Journal of Micromechanics and Microengineering, vol. 31, no. 12, 2021, p.125008 (11pp).

International Conference (selected)

1. H. Toshiyoshi, "Design and Test for MEMS Vibrational Energy Harvesters," 35th International Conference on Microelectronic Test Structures (ICMTS), March 27, 2023, The University of Tokyo, Tokyo, Japan. (Tutorial)
2. Mohammed Saad Khan, Changdae Keun, Yi Xiao, Vivek Anand Menon, Keiji Isamoto, Nobuhiko Nishiyama, and Hiroshi Toshiyoshi, "Compact low-cost solution for wavelength sensitive applications with micro-machined tunable VCSEL," in Proc. 2022 IEEE International Conference on Optical MEMS and Nanophotonics (OMN 2022), Sept. 12-15, 2022, online.
3. Refaldi Intri Dwi Putra, Takahiro Ozawa, Hiroaki Honma, Hiroshi Toshiyoshi, and Katsuyuki Fukutani, "Effects of Gas Adsorption and Ions on the Reliability of Potassium-ion SiO2 Electret," in Proc. 22nd Int. Vacuum Congress (IVC-22), Sept. 11-16, 2022, Sapporo Convention Center, Sapporo, Japan, Tue-PO1C-30.
4. Hiroshi Toshiyoshi, "MEMS Vibrational Energy Harvester for IoT Sensors," in Proc. 2022 JSME-IIP/ASME-ISPS Joint Conference on Micromechatronics for Information and Precision Equipment (MIPE2022), Aug. 28-31, 2022, Nagoya University, Nagoya, Japan, D3-2-01 (1p). (Hybrid) (Invited Keynote)
5. Yuto Akai, Hiroaki Honma, and Hiroshi Toshiyoshi, "Vibrational MEMS Energy Harvester Capable of Monitoring Phase State Variables," in Proc. Asia-Pacific Conference of Transducers and Micro-Nano Technology 2022 (APCOT 2022), May 29 - June 1, Shanghai, China (Hybrid all online)
6. Masahide Goto, Yuki Honda, Masakazu Nanba, Yoshinori Iguchi, Takuya Saraya, Masaharu Kobayashi, Eiji Higurashi, Hiroshi Toshiyoshi, and Toshiro Hiramoto, "3-Layer Stacking Technology with Pixel-Wise Interconnections for Image Sensors using Hybrid Bonding of Silicon-on-Insulator Wafers Mediated by Thin Si Layers," 2022 IEEE 72nd Electronic Components and Technology Conference (ECTC 2022), May 30 - June 3, 2022, San Diego, CA, USA.
7. Hiroaki Honma, Shota Harada, and Hiroshi Toshiyoshi, "Demonstration of production of pull-in cancellation voltage generated by electret-based vibrational energy harvester and Cockcroft-Walton voltage multiplier," in Proc. 17th IEEE International Conference on Nano/Micro Engineered & Molecular Systems (IEEE-NEMS 2022), April 14-17, 2022, Chang Gung University, Taoyuan, Taiwan.
8. Hiroshi Toshiyoshi, "Silicon Oxide Electret as a Power Generation Material," in Proc. 17th IEEE International Conference on Nano/Micro Engineered & Molecular Systems (IEEE-NEMS 2022), April 14-17, 2022, Chang Gung University, Taoyuan, Taiwan (ONLNE). (Invited Keynote)

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A normally off time-of-event logging system triggered by a battery-less sensor☆

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ABSTRACT

This paper presents a novel time-logging system using a real time clock (RTC) powered by a vibrational energy harvester, triggered by a piezoelectric polyvinylidene fluoride (PVDF) film as an event-driven sensor. The power-management circuit normally remains in sleep mode to cut off the power supply to the micro-processor and the ferroelectric random access memory (FRAM), while the RTC alone is continuously powered by a battery to keep the time for seamless operation, thereby lowering the standby power consumption to as small as 231 nW. The power management circuit for a PVDF power generator can supply a load current upwards of 8 mA at 3.3 V_{dc} for the external circuit. Time-of-event recording is performed with energy consumption of 568 μJ per event.

1. Introduction

WIRELESS sensor network has attracted increasing attention to avoid accidents such as corruption of infrastructures, crimes, and disasters, which endanger people in daily life. For such a purpose, wireless nodes are used to collect tremendous amounts of data and upload them to the clouds through the local networks (Wi-Fi) or the cellular network connectivity. The wireless nodes are usually programmed to check the conditions of objects by tracking the variations of the physical values using aforementioned data acquisition system.

As of data acquisition, the time of event is essential to associate the data with the timestamp of the event for subsequent big data analysis, and therefore a timer should be included in every wireless sensor module. To date, three approaches are known to keep time, including atomic clocks [1,2], the global positioning system (GPS), and real time clock [3,4] (RTC). The atomic clock and the GPS signal have high accuracy because they are based on the electron transition frequency that is inherent to the nature of atoms. They are, however, incompatible with wireless sensor nodes due to the large power consumption of a few mW or more. In contrast to this, typical RTC has power consumption as small as a few hundreds of nW, which could be managed by energy harvesters for environmental energy sources [5–11]. The precision of RTC is not as high as those of the atomic clocks or the GPS signals; nevertheless, the error is as small as 30 s a year [12], which is negligibly small for certain applications of relatively long time constant such as tracking service for delivery, agriculture monitoring, and detection of car accident as shown in Fig. 1. Furthermore, RTC is developed by the silicon technology, which is compatible with the silicon-base microelectromechanical systems (MEMS) process for potential integration of RTCs with energy harvesters in the coming years.

The output of energy harvester today is not sufficient to operate wireless sensor module for IoT [13–18], and thus it is required to develop a wireless sensor module of extremely low power consumption. Although such modules are expected to monitor objects to detect errors or abnormality, sleep mode is dominant in the whole module lifetime. A normally-off module, therefore, is usually used to cut off power consumption under the sleep mode. Those of modules, however, would overlook crucial signs which lead to catastrophic accidents.

For this technical issue, we adapted an energy harvester as a sensor to activate the IoT module which does not miss to report an event. In this paper, we discuss on the circuit architecture to imprint the time stamp of an RTC into the memory by using a vibrational energy harvester as an event-driven switch (EDS). The developed system has a function to detect the external events which should not be overlooked by the

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Effect of hydrogen atoms on potassium-ion electrets used in vibration-powered generators

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A R T I C L E I N F O

Keywords:
First-principles calculation
Vibration-powered generator
SiO_2
Energy harvesting
Electret

A B S T R A C T

Potassium-ion electrets, which are mainly composed of amorphous silica and potassium atoms, can accumulate negative charges and are expected to be useful for future vibration powered generators. According to previous research, the origin of the accumulation of negative charges was revealed to be related to the presence of SiO_5 structures in amorphous silica. In general, Si–O bonds are so stable that potassium-ion electrets are expected to retain a high electric potential longer than previously studied materials and are therefore being studied intensively for practical use. However, charge decay phenomena were observed in previous experiments, and this presents serious problems. In this study, we focus on the role of hydrogen which is considered to be one of the origins of charge decay and have investigated the behavior of hydrogen in potassium-ion electrets and the mechanism of the charge decay using first-principles calculations. As a result, we found that hydrogen atoms can create a positively charged characteristic local structure or break SiO_5 structures and therefore cause degradation of the negative charge storing ability of potassium-ion electrets.

1. Introduction

The realization of Internet of things (IoT) for our future convenience has fueled the development of innovative technologies such as artificial intelligence and precision sensors. However, the demand of autonomous power sources which supply the electric power to IoT devices is also increasing. As a result, energy harvesting technology [1–4] has recently been paid a lot of attention. This collects many kinds of energies that exist in the natural environment, such as light, heat, and vibration and converts them into electrical power. Amongst all the possibilities, vibration powered generators are expected to be especially useful for IoT devices since they operate independently of daily temperature and natural phenomena changes. Vibration is generated in many places, such as movement of humans, of roads or bridges, of vehicles and machines, so the range of application of vibration powered generators is considered to be very wide [5].

Micro electro-mechanical systems (MEMS) vibration-powered generation devices incorporating potassium-ion electrets have been developed recently and are expected to be one of the most promising vibrational energy harvesting devices. Electrets are materials which can accumulate charge and therefore develop an electric field. Potassium-ion electrets [6–11] are composed of amorphous silica and potassium atoms and can accumulate negative charges. The origin of accumulation of negative charges is understood to be due to the presence of SiO_5 structures in the amorphous silica [12]. Si atoms in amorphous silica usually have four Si–O bonds and form a tetrahedral structure. However, in potassium-ion electrets, some Si atoms have five Si–O bonds and accumulate negative charges, due to the capture of electrons from potassium atoms. Si–O bonds are so strong that the SiO_5 structures are highly stable and expected to accumulate charges semi-permanently. In fact, it was estimated that the lifetime of potassium-ion electrets is about 400 years [13]. Because of their useful features, potassium-ion electrets are expected to be used in maintenance-free electrostatic-type vibration-powered generators in the future.

However, such electrets have yet to be put to widespread use. The main problem is charge decay of the electret. We have recently

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Improvement of the reliability of potassium-ion electrets thorough an additional oxidation process

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ABSTRACT

Potassium-ion electrets, which mainly consist of amorphous silica and permanently store negative charge, are vital elements of small autonomous vibration-powered generators. However, negative charge decay is sometimes observed to cause issues for applications. In this study, we propose an improved guideline for the fabrication of potassium-ion electrets by first-principles calculations based on density functional theory, which shows that yield reduction can be suppressed by additional oxidation. Moreover, we experimentally confirm that the additional oxidation step effectively extends the lifetime of potassium-ion electrets.

Extracting electric power from energy that exists thinly and widely in the natural environment is called energy harvesting. At present, research on energy harvesting technologies that extract and effectively utilize electric power from various naturally occurring environmental elements, such as solar, wind, and geothermal power, is being actively conducted around the world.[1–4] It has attracted a great deal of attention recently because it can contribute to the realization of the Internet of Things (IoT) through its utilization in maintenance-free autonomous power generation devices. Such energy harvesting devices tend to have very small maximum power outputs and have, therefore, had limited use in conventional electronics. Additionally, the outputs of some energy harvesting technologies depend on weather conditions and are very unstable as autonomous power devices. However, the range of applications for energy harvesting devices has expanded due to the innovation of low power device technologies and vibration-powered generator technology, which is one of the energy harvesting technologies expected to efficiently generate electricity because it is neither dependent on weather conditions nor on the time of day.[5]

Micro-electromechanical systems (MEMS) vibrational power generation devices incorporating potassium-ion electrets have been developed recently and are expected to be one of the most promising energy harvesting devices.[6–11] The device is based on a SiO_2 thin film, which is charged via potassium ions. Because the SiO_5 structure, which has five strong Si–O bonds, can accumulate negative charges, potassium-ion electrets are expected to be important materials for maintenance-free devices.[12] When the potassium-ion electret and an electrical conductive plate are placed in parallel, an induction current is obtained by moving the plate through vibration, making it possible to realize a maintenance-free power generation device that can operate semi-permanently. However, fabrication issues have been reported, which result in the potassium-ion electrets not charging effectively despite using similar fabrication procedures, resulting in a yield reduction.[12] It has been suggested that this issue is related to Si–Si bonds in a-SiO_2.[12] In this paper, we propose a guideline for the fabrication of potassium-ion electrets to improve the yield.

We used first-principles calculations based on the density functional theory. All the calculations were performed using the Vienna *Ab initio* Simulation Package (VASP) code.[13–16] We used the Perdew–Burke–Ernzerhof (PBE) exchange–correlation functional[17] for molecular dynamics (MD) calculations and structural optimization calculations, and the Heyd, Scuseria, and Ernzerhof (HSE) exchange–correlation functional[18] to better describe the energy levels including bandgaps. We set the fraction of exact exchange and the

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Paper

Double-Deck MEMS Electrostatic Vibrational Energy Harvester with Airborne Interconnection

Hiroaki Honma*[a] Member, Hiroshi Toshiyoshi* Senior Member

(Manuscript received Feb. 2, 2022, revised March 23, 2022)

Lowering internal impedance, i.e., stray capacitance and resistance, is essential to enhance the output power of microelectromechanical system (MEMS) vibrational energy harvesters particularly when the structure is made of a silicon-on-insulator (SOI) wafer which has a distributed stray capacitance across the buried oxide (BOX) layer. Instead of using silicon as a material for electrical interconnection, we used an out-of-plane aluminum bonding wire to complete the on-chip electrical interconnection between the contact pads on the chip frame and the suspended movable electrodes. Compared with the conventional design using silicon-based interconnections, the maximum deliverable power has been enhanced almost six-fold because of the reduced stray capacitance between the SOI layer and the substrate, as well as reduced internal electrical resistance. The mechanical stability of the bonded wire during the excitation vibration has also been experimentally confirmed. A typical power density of 763 μW/cm^3 is obtained when excited by 0.5 G (1 G = 9.8 m/s^2) sinusoidal vibrations at 157 Hz.

Keywords : MEMS, energy harvester, electret, environmental vibration, double-deck

1. Introduction

Compared with the rapid miniaturization of integrated circuits, energy source modules still remain too bulky to construct power-autonomous IoT (internet of things) microelectronics. Considering the around-the-clock availability of unharnessed environmental power, a multi-modal combination of energy sources is preferred, including solar cells[(1)], mechano-electric transducers[(2)], thermoelectric transducers[(3)], secondary batteries and capacitors[(4)]. Ambient vibrations in particular are ubiquitously available in every man-made structure and piece of machinery, and a variety of mechano-electric conversion mechanisms have been studied including electromagnetic[(5)], piezoelectric[(6)], and electrostatic induction[(7)]. In our recent work, a high-density electret material has been developed for a MEMS (microelectromechanical systems) vibrational energy harvester with a large power density of 763 μW/cm^3[(8)].

The power density of electrostatic energy harvester could be further improved by enhancing the electret density, adjusting the mechanical resonance frequency exactly to that of the incoming vibrations, and fulfilling the impedance-matching condition between the harvester and an external load[(9)]. From a practical point of view, the device size should also be minimized to facilitate system-in-package implementation for IoT devices in a limited volume. A double-deck MEMS structure has been thus developed for energy harvesters, using both the front and back sides of a silicon-on-insulator (SOI) wafer to respectively form the electrodes and suspension, as reported elsewhere[(10)]. The device footprint has become almost half the size of the conventional design in which the electrodes and suspension were flattened in a single layer[(8)]. However, this size reduction inevitably caused an associated increase in parasitic capacitance that unintentionally increased the reactive power, thereby lowering the net output.

In this paper, we present a method to avoid this side effect in the double-deck structured MEMS vibrational energy harvester. A simple airborne bonding wire is used for electrical interconnection, which significantly mitigates collateral stray capacitance. In the following section, we discuss the device structure and equivalent circuit model. We also present experimentally confirmed harvester performance. This work includes experimental results that were published in an international conference[(11)].

2. Electrostatic Energy Harvester

2.1 Operating Principle Figure 1 illustrates the operating principle of the vibrational energy harvester based on electrostatic induction. A pair of interdigitated comb electrodes made of single crystalline silicon is set with a tiny air gap between them as shown in Fig. 1 (a); the surface of the electrode is coated with a conformal layer of silicon oxide that has been permanently charged to be an electret. As reported elsewhere, silicon oxide with impurity ions can be turned into an electret after being electrically biased in a large electrostatic field (5 kV/cm) at high temperature (>500 °C)[(12)]. In this work, the silicon oxide film on the fixed electrode is negatively charged with respect to the movable electrode, which is electrically grounded.

The electrostatic flux arising from the negatively charged electret is usually coupled with a hole in the underlying fixed electrode. When the movable electrode is pushed into the gap by an external force as shown in Fig. 1 (b), a part of the flux is redirected to the holes in the movable electrode due to the increase in overlap between the two electrodes, thereby releasing the electrons that are transferred to the fixed electrode through an external resistor. Therefore, the mechanical power from the external force is converted into electrical power, which is consumed as Joule heat. In the case of a practical energy harvester, the gained energy is stored in a capacitor until discharged to the subsequent electronics.

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MEMS Switching Voltage Regulator Using a Normally-On Electret Relay

Mizuki Morikawa, Yasushi Shibata, Hiroshi Toshiyoshi, *Member, IEEE*, and Gen Hashiguchi

***Abstract*— We report on a DC/DC step-down switching regulator using a microelectromechanical systems (MEMS) switch for the first time. The output voltage developed in a storage capacitor is used as a feedback control voltage for the electrostatic actuator that drives a micromechanical contact switch inserted between the rectifier and the capacitor. The switch is configured to be normally-on owing to the electrostatic potential remaining in the electret film on the electrodes. When the output voltage exceeds and the feedback control voltage cancels the built-in potential of the electret, the differential voltage acting on the actuator becomes small and the contact is opened, thereby decoupling the rectifier from the storage capacitor. When the output voltage becomes low, on the other hand, the net differential voltage becomes large and the contact is closed again, thereby reconnecting the rectifier to the capacitor and recovering the voltage. Fast repetition of this sequence maintains the DC voltage of the storage capacitor at a constant level. For demonstration, an electret film that is pre-charted to 240 V is used to regulate the output voltage in a range between 12 V and 14 V. Different from the switching regulators using transistors, the MEMS regulator does not require any bias current for operation, being suitable as a low-power voltage regulator for environmental energy harvesters. [2021-0220]**

***Index Terms*— MEMS, voltage switching regulator, electret, comb-drive actuator, vibrational energy harvester.**

I. Introduction

RECENT R&D of microelectromechanical systems (MEMS) energy harvesters have made rapid strides, and a normalized power density (NPD) as high as 30.9 mW/cm^3/G^2 (1 G is 9.8 m/s^2) has been realized using a comb-electrode mechanism that is electrically biased with a static field of a built-in electret [1]. Energy harvesters of electrostatic type generally produce voltages high enough to overcome the PN-junction barrier of silicon diode rectifier, and the harvested energy can be stored in a capacitor building a relatively large dc voltage. For instance, a capacitor of 44 μF was charged to 14.7 V in 90 minutes using the output voltage of an energy harvester driven by the non-sinusoidal random vibrations of a highway viaduct [2].

Nevertheless, the output current of the harvester is usually limited to a sub-μA $\sim$ μA level (118 nA in [2]) because of the large output impedance, narrowing the range of applications to low power electronics only. For more practical use, the energy stored in the capacitor would be used intermittently, repeating the charge and discharge cycles with a power management circuit. Such a circuit should be designed to regulate the output voltage to a convenient level, which is typically 3 to 5 V for the subsequent circuits, while maximizing the supply current and minimizing the stand-by current.

Comprehensive study on power management circuits for energy harvesters was reported by Szarka *et al.* in 2012 [3], in which the-state-of-the-art circuit elements were introduced. In 2018, Galayko *et al.* reported an updated review on the electrostatic and piezoelectric vibrational energy harvesters and their power conditioning electronics [4]; the same authors also developed a power management circuit inspired from the Bennet's voltage doubler [5], [6]. CMOS (complementary metal-oxide-semiconductor) power management circuits are under intensive study to maximize the retrievable power from small kinetic energy [7]–[14]. Some have already been commercialized as PICs (peripheral interface circuits) that operate at a very small stand-by current of only 1 nA [15] as released, for instance, from Microchip Technology, Inc. As for a timer IC (integrated circuit) that is indispensable to control the timing of operation, a low-power product operating at 35 nA is commercially available from Texas Instruments, Inc., [16]. However, no solution has been reported to reduce the operation current of the voltage regulator ICs to a level lower than 100 nA so that voltage regulation would be manageable within the power budget of harvested energy.

To solve this problem, we propose a method to use a MEMS electrostatic switch in place of a semiconductor transistor in the electronic voltage regulator circuit. Typical voltage regulator is constructed using a MOS (metal-oxide-semiconductor) switch and a voltage comparator for feedback control [8], [9]. In contrast to this, we replace the MOS switch and the comparator respectively with a MEMS contact switch and the electrostatic actuator's behavior that is governed by the difference of voltages applied to the electrode pair. As illustrated in Fig. 1(a), a MEMS contact switch of a normally-on type is inserted between the rectifier's output and the storage capacitor as in the same topology of the switching regulator composed of MOS transistors shown in Fig. 1(b), where the storage voltage is referenced by the switch element for feedback control. One

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Digital Object Identifier 10.1109/JMEMS.2022.3151909

KAWAKATSU Laboratory

Current Research Activities 2021-2022

Kawakatsu Laboratory

1. Research Topics

1.1 Colour Atomic Force Microscopy with Chemical Contrast

In this project, the AFM cantilever is oscillated at 2 MHz, and the tip-sample distance is modulated at 1kHz. By employing such modulation techniques, the potential landscape is detected as the frequency shift of the cantilever. Three effective Morse parameters are calculated per pixel to best curve fit the measured values. The work was published in 2017 on APL and was selected as the editor's pick. In 2020, discussions on the control scheme as well as results on imaging of solder was published in JJAP.

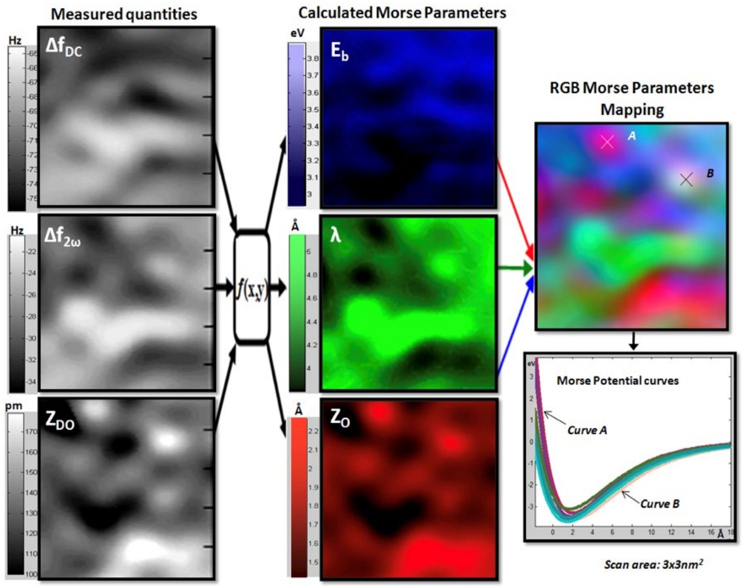
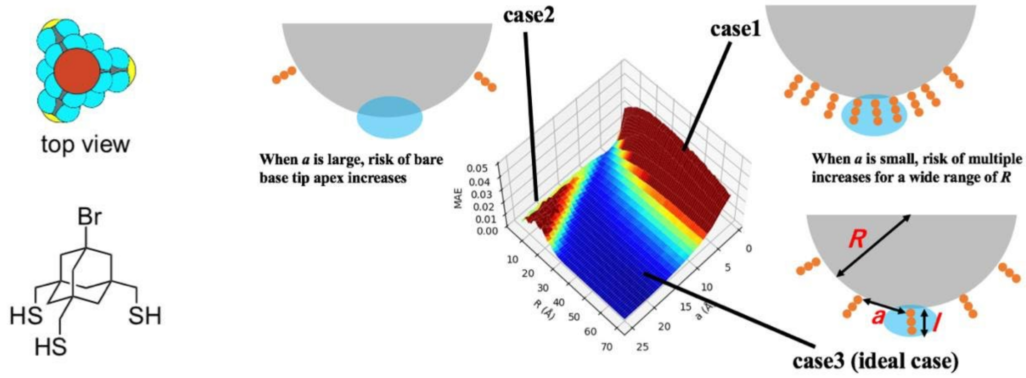


Figure 1. Effective Morse Parameters represented in RGB obtained from modulation of the tip sample distance. The sample was quenched Si(111). Cantilever Oscillation 2MHz.

1.2 Theoretical considerations for Functionalisation of the AFM tip apex with thiol tripod structured molecules

Adamanitine-3-thiol (ATT) molecules are a new candidate for functionalizing AFM tip apexes due to their size, stability and their chemical bonding to the AFM tip. The molecules are synthesized by Professor T. Kitagawa of Mie University. Two projects are running under this topic. One is the macroscopic measurement of the range of stable operation with the ATT tips. Second is the simulation on tips functionalized with the ATTs when used for atomic force microscopy. For the first experiment, quartz oscillators with gold electrodes are used for self-assembled monolayer growth of ATTs. The frequency of oscillation of the quartz oscillators, which lie in the range of 1MHz to 10MHz are monitored as a function



(a) (b)

Figure 2 (a) Bromadamantane-3thiol, and (b) Average ***MAE*** (mean absolute error) for different values of ***R*** and ***a***. case 3 corresponds to the ideal case, giving results closer to imaging with a single atom tip. The blue area plays the most important role in AFM imaging.

of temperature in air and in vacuum to assess the detachment temperature of the molecules from the gold surface. As for optimization of base tip radius and spacing between the molecules, and the height of the molecules, 100 to 1000 functionalised tips randomly generated for simulation were subject to force curve studies and image quality compared to an ideal floating single atom "probe". The results to be published.

<u>1.3 Conductive Polymer Dehydrant (Joint research with Tohoku University)</u>
Conductive polymers have been known to show absorption of humidity in air. This project is a collaboration with Professor Hikaru Kobayashi of Tohoku University. We focused on various design for removing humidity from air, as well as improving conductance of the functional polymers and its deterioration due to current, heat and exposure to light. The conductive property was used to heat the polymer with Joule's heat to dehydrate the polymer for the next dehydration cycle. Honeycomb scaffolding covered with the conductive polymer was supported by mechanical oscillators oscillating around 10 to 30 Hz to measure accumulation of water through frequency shift. Mass resolution in the 10 mg order was confirmed. The method provided a simple in-process scheme to monitor breathing of humidity by the device.

<u>1.4 High slew rate circuit for ultra-high rigidity friction drives.</u>
A new circuit was developed to allow the use of large surface area piezo-elements for unprecedented high rigidity and stability in SPM heads. UHV and ambient condition AFMs were implemented based on this driving principle. The circuit uses discharge of accumulated charge of the piezo element by short-circuiting with a transistor. High voltage tolerance and high current capacity were demonstrated, giving unprecedented performance compared to commercial linear high-voltage amplifiers(D. Kobayashi and H. Kawakatsu, JJAP, 2020).

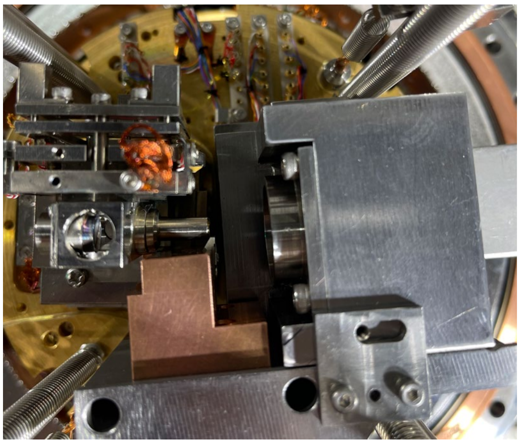


Figure 3. Improved Colour AFM head for higher rigidity and stability. From left to right, objective lens, cantilever holder, sample stage and coarse positionner.

1.5 Improved colour AFM head for higher rigidity and stability

The AFM head used for Colour Atomic Force Microscopy was redesigned and rebuilt for higher rigidity and stability. Friction drives with large surface area were driven by the high-slew rate driving circuit developed in our laboratory (D. Kobayashi and H. Kawakatsu, JJAP 2020)

1.6 Listening to sperm activity with a high-sensitivity in-liquid electret microphone

A high-sensitivity in-liquid electret microphone with a diameter of ca. 5 mm was placed in the double suspended vibration isolation table. It was shown that medium with and without the sperms showed different acoustic spectra(Fig.4). The power intensity was higher for samples with the sperms. It was also shown that the centre of acoustic spectrum distribution shifted towards the higher frequency as the incubation of the sperms progressed. This may be related to published results where the sperm tail becomes more motile following incubation.

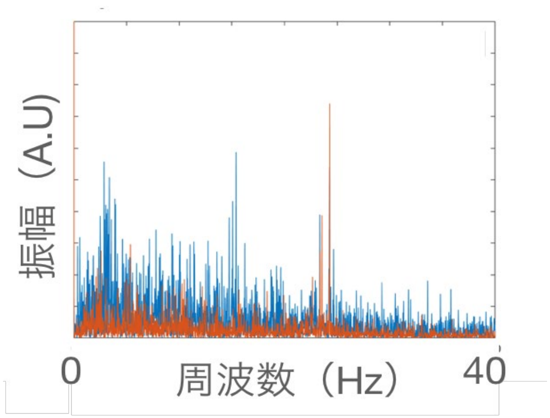


Fig 4. Acoustic spectrum of media with(blue line) and without(red line) spermatozoa. Sperm concentration not controlled.

2. Research Achievements

2.1 Number of original journal papers: 0

2.2 International conference: 2 (including 2 invited presentations)

2.3 Domestic conference: 3

2.4 Number of patents: 0

3. List of awards

4. Research Grants

4.1 Total number of research grants: 3

4.2 Number of collaboration research with industry: 5 (not excluding non-Grant based collaborations)

5. Education

5.1 Number of Ph.D. students :0

5.2 Number of master students: 4

5.3 Number of other students: 2

6. Publication list

Conference Presentations

1)Study on regeneration of a conductive polymer dehydrant by Joule heating (in Japanese, tentative title), Sirasu K.Kobayashi H.,Itoh K.,Yang S., Kobayashi D., and Kawakatsu H., in Proc. of Architectural Inst. of Jpn., 1867-1868, 2022-09-05.

2)Measurement of absorption and desorption characteristics of water of a conductive polymer with a mechanical vibrator type mass sensor(in Japanese, tentative title), Nishiyama H., Kobayashi H., Kawakatsu H., Kobayashi D., and Kanai D., in Proc. of The society of heating, air-conditioning and sanitary engineers of Japan, Tohoku division,12 41-45, 2023-03-03.

3)Development of a dehumidifier using PEDOT/PSS with a regeneration function by Joule Heating -Optimisation of design and control- (in Japanese, tentative title), Higuchi Y., Kobayashi H., Kawakatsu H., Kobayashi D., and Kanai D., in Proc. Of The society of heating, air-conditioning and sanitary engineers of Japan, Tohoku division,,12 43-46, 2023-03-03

KIM Laboratory

Current Research Activities (2022 April-2023 March)

B.J. KIM Laboratory

1. Research Topics

Our research goals are to build smart nanosystems and integrate nanoscale components in micro sensors, in particular for environment, bio-sensing, through both bottom-up and top-down approaches. We focus on interdisciplinary research about local “bottom-up” surface modification using functional self-assembled monolayers and “top-down” approaches for micro/nano patterning technologies. Based on these studies on nano/micro components systems for the fabrication of novel nano devices, we investigate to develop various micro sensors for biological applications, health care as well as environmental monitoring.

In the transdermal drug delivery methods, **the microneedle-mediated drug delivery system (DDS)** has been developed to replace the hypodermic injection-mediated DDS, to provide painless self-administration of biological drug with patient friendly manner. Dissoluble microneedles are attracting much attentions as it has several advantages such as no needle-related risks. We have developed new fabrication method for biodegradable microneedles patches, which is different with the conventional fabrication ones, such as stepwise casting method. We anticipate that shadow mask assisted drawing lithography as well as 3D printing based novel mass fabrication methods, will be suitable to improve the fabrication throughput of dissoluble microneedle for new generation of drug delivery system.

On the other hands, we observed the permeability of several commercially-available microneedle patches for cosmetic purposes to human skin and investigated methods of evaluating permeability to the stratum corneum, and its’ degree of pain.

1.1 Dissoluble Microneedle patch for transdermal drug delivery systems

The transdermal drug delivery has experienced several-generation revolutions: From the transdermal delivery of small, lipophilic, low-dose drug to the delivery system using chemical enhancer, non-cavitational ultrasound and iontophoresis. With the development of the micro-scale engineering, microneedles show the potential to be the next generation delivery system. The microneedle mediated drug delivery system has been developed to provide painless self-administration of biological drug with patient friendly manner. Especially, dissolving microneedles, which deliver the target drugs as the drug-loaded microneedle dissolves into the skin, have been spotlighted recently. We investigate a novel fabrication method to achieve the user-friendliest, low-cost, and safest way for dissoluble microneedle patches with vaccine delivery and several medical treatments.

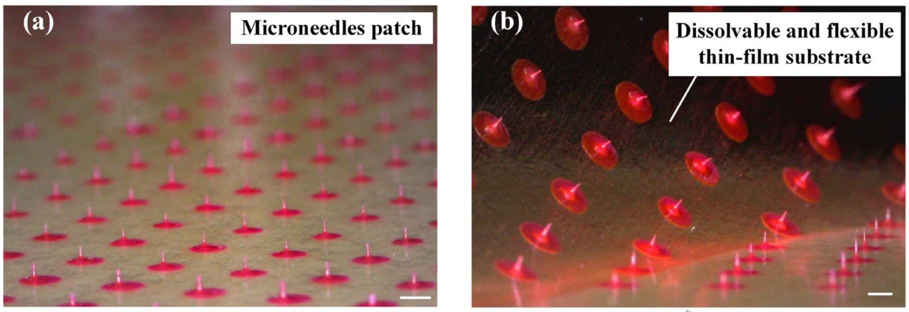


Fig. 1: *Dissoluble microneedles of Hyaluronic acid with red dye for transdermal drug delivery.*

1.2 Porous Microneedle for Self-testing diagnostic Bio-sensing

The porous microneedles are expected to have great potential for diagnostic application due to their ability to penetrate human skin painlessly and extract interstitial fluid (ISF) by capillary action. However, the prior approach about porous microneedles had not directly integrated sensor system as an application due to the small amount of sample porous microneedle can absorb. Here, we fabricated porous microneedle on a paper substrate to develop a novel platform for direct integration of sensors.

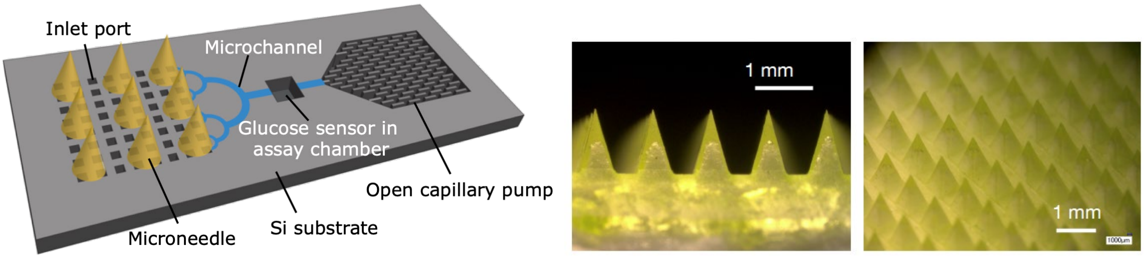


Fig.2: *Schematic of minimally invasive glucose monitoring system (left) integrated with an array of PDMS porous microneedles (right) in a microfluidic chip.*

ISF is a promising bio-sample since it contains a wide range of common biomarkers to blood such as glucose. In order to utilize ISF for continuous healthcare monitoring, ISF sampling is a key technology. However, the conventional ISF sampling technologies such as microdialysis have several drawbacks including highly invasive operation and limited measurement methodologies. Among the MNs, porous MNs have advantages such as applicability to fluidic systems and to biocompatible materials. Although the porous MNs were applied to ISF sampling, continuous ISF sampling has not been realized. For this, a new type of porous MN should be developed to address the challenges of mechanical and fluidic requirements for successful insertion into the skin and ISF extraction continuously. Furthermore, the microfluidic chip should also be realized to interface the porous MNs and realize a continuous flow of ISF, which is ideally at a flow rate of 0.08 μL/min.

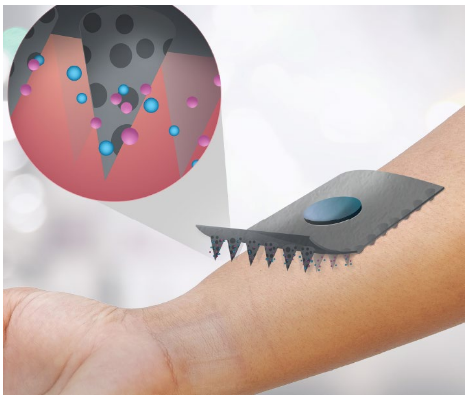


Fig.3*: Porous microneedle-on-paper device with a simple colorimetric sensor can painlessly measure glucose levels from interstitial fluid in the skin.*

One of the core targets is employing the micro-electro-mechanical systems (MEMS) fabrication technologies to the bio-degradable materials, then realizing specific structures, such as free-standing membrane, ultra-thin transparent film, etc. Therefore, our aim is to employ micro-nano technology to accurately control and deeply investigate the bio-degradable materials at micro/nano level, and develop novel materials for multiple application fields, such as biomedical field and energy harvesting.

As the power sources of wireless sensors, batteries are mainly utilized until now. These days, devices of energy harvesting become more focused. **Energy harvesting** is a methodology of scavenging power from ambient energy sources such as solar, thermal and vibration without the need for batteries.

In 2012, a novel energy harvesting approach was proposed and named TENG (**Triboelectric Nano Generator**), which is based on the combination of electrification effect and electrostatic induction. Due to remarkable properties, such as high-performance and the use of environmentally friendly materials, TENG has raised increasing interest. Several techniques have been developed to enhance the power density, including structural optimization, operation mechanism, surface texturing and hybrid TENG.

Herein, we propose a very simple and cost-efficient approach to fabricate high-performance TENG based on paper and graphite pencil. Moreover, we propose a novel device, which can utilize ambient vibration with wideband and low frequencies. Due to high stiffness of piezo electric energy harvesters, resonant frequency of the piezo electric energy harvesters tends to be much higher than ambient vibrational frequency. To solve these problems, the proposed energy-harvesting device is to utilize a stochastic resonance.

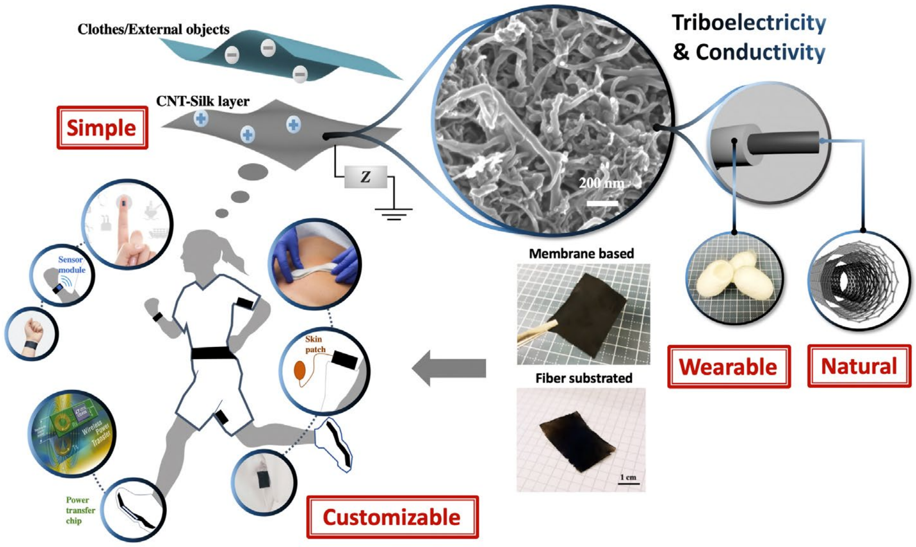


***Fig. 4**: Wearable TENG device of the electro-sprayed silk fibroin fiber mixed with carbon nanotubes.*

This high-output energy harvester shows the attractive potential in the integrated flexible electronics and wearable device.

2. Research Achievements (2022年度中)

2.1 Number of original journal papers: 6 (113 total)

2.2 International conference: 10 (227 total),

2.3 Domestic conference: 4 (211 total)

2.4 Number of patents: 3 (17 total)

3. Research Grants

3.1 Total number of research grants: 7 (2021-2022)

3.2 Number of collaboration research with industries: 5

3.3 List of major research grants (serving as Principal Investigator)

- JSPS Core-to-Core A, JETMeE project, AMED 橋渡し研究戦略プログラム.

4. Education

4.1 Number of Ph.D. students (including current students): 13(4)

4.2 Number of master students (including current students): 47 (5)

4.3 Number of other students: 37 (including current students 2)

5. Publication list

Journal Paper (2021-2022) *attached

1*. Leilei Bao, Jongho Park, Boyu Qin, and Beomjoon Kim: Anti-SARS-CoV-2 IgM/IgG antibodies detection using a patch sensor containing porous microneedles and a paper-based immunoassay, ***Scientific Reports***, 12:10693, 2022 (https://doi.org/10.1038/s41598-022-14725-6)

2*. Xiaobin Wu, Jongho Park, Siu Yu A. Chow, Maria Carmelita Z. Kasuya, Yoshiho Ikeuchi, and Beomjoon Kim: Localised light delivery on melanoma cells using optical microneedles, ***Biomedical Optics Express***, Vol. 13, Issue 2, pp. 1045-1060, 2022 (https://doi.org/10.1364/BOE.450456)

3*. Kai Takeuchi, Nobuyuki Takama, Kirti Sharma, Oliver Paul, Patrick Ruther, Tadatomo Suga, Beomjoon Kim: Microfluidic chip connected to porous microneedle array for continuous ISF sampling, ***Drug Delivery and Translational Research***, 12, pp. 435-443, 2022 (https://doi.org/10.1007/s13346-021-01050-0)

4*. Leilei Bao, Jongho Park, Gwenael Bonfante, and Beomjoon Kim: Recent advances in porous microneedles: materials, fabrication, and transdermal applications, ***Drug Delivery and Translational Research***, 12, pp. 395-414, 2021

5*. Libo Wu, Jongho Park, Yuto Kamaki, and Beomjoon Kim: Optimisation of fused deposition modelling based fabrication process for polylactic acid microneedles, Springer Nature, ***Microsystems & Nanoengineering***, 7 (58), 2021

6. Dan-Liang Wen, De-Heng Sun, Peng Huang, Wen Huang, Meng Su, Ya Wang, Meng-Di Han, Beomjoon Kim, Jürgen Brugger, Hai-Xia Zhang, and Xiaosheng Zhang: Recent Progress on Silk Fibroin-Based Flexible Electronics, ***Microsystems & Nanoengineering***, 7, 35, 2021 (https://doi.org/10.1038/s41378-021-00261-2)

7. Hai-Tao Deng, Xin-Ran Zhang, Zhi-Yong Wang, Dan-Liang Wen, Yan-Yuan Ba, Beomjoon Kim, Meng-Di Han, Hai-Xia Zhang, Xiao-Sheng Zhang: Super-stretchable multi-sensing triboelectric nanogenerator based on liquid conductive composite, ***Nano Energy***, Vol.83, 105823, 2021 (https://doi.org/10.1016/j.nanoen.2021.105823)

International Conference (2022 y, selected)

1. Dominique Decanini, Abdelmounaim Harouri, Beomjoon Kim, Yoshio Mita, Gilgueng Hwang: Manufacturing of magnetically reconfigurable 3D printed micro fractal pipette array, ***French symposium on emerging Technologies for Micro-Nanofabrication (JNTE 2022)***, p. 29, Besancon, France Nov. 28- Dec. 2, 2022

2. Heyi Jing, Boyu Qin, Leilei Bao, Jongho Park and Beomjoon Kim: Porous microneedles integrated paper sensor for cholesterol measurement, ***The 26th. International Conference on Miniaturized Systems for Chemistry and Life Sciences, (MicroTAS 2022)***, Hangzhou China (hybrid), pp. 1133-1134, W150.f, 23-27 October, 2022 (poster)

3. Jongho Park, and Beomjoon Kim: Biotagging method using microneedle arrays for individual identification of animals, ***The 19th. International Conference on Precision Engineering (ICPE 2022 in Nara)***, Nara, Japan, Nov.28-Dec. 2, 2022

4. Yosuke Koma, Yuko Tsuruma, Jongho Park, Shigenori Aoki, Shinya Takyu, Beomjoon Kim: Development of a simple glucose sensor patch using low melting-point polymer's porous microneedles for pre-diabetic patients, ***11th. IEEE CPMT Symposium Japan 2022* (ICSJ 2022)**, Kyoto Univ. Clock Tower Centennial Hall, Japan, Proceeding of ICSJ 2022, pp. 115-118, November 9-11, 2022 (oral)

5. Boyu Qin, Jongho Park, Heyi Jing, Leilei Bao, Beomjoon Kim: Porosity control of polylactic acid porous microneedles using microfluidic technology, ***11th. IEEE CPMT Symposium Japan 2022* (ICSJ 2022)**, Kyoto Univ. Clock Tower Centennial Hall, Japan, Proceeding of ICSJ 2022, pp.127-130, November 9-11, 2022 (oral)

6. Beomjoon Kim: Bio molecular Needling System and healthcare, ***International Conference on Manipulations, Automation and Robotics at Small Scales (MARSS 2022)***, Toronto, Canada, July 25-29, 2022 (Plenary talk)

7. Xiaobin Wu, Jongho Park, and Beomjoon Kim: An optical Microneedles for Localized Light Therapy on Melanoma cells, ***The 35th. IEEE International Conference on Micro Electro Mechanical Systems (IEEE MEMS 2022)***, Hybrid Conference, Tokyo, Japan, Proc. of MEMS 2022, pp. 313-316, Jan. 9-13, 2022 (poster presentation)

8. Kai Takeuchi, Beomjoon Kim, Tadamoto Suga: Prolongation of Surface Activation Effect using a Self-Assembled Monolayer for Low Temperature Bonding of Au, ***the IEEE 72nd Electronic Components and Technology Conference (ECTC)***, G01-1631, San Diego, California, USA, May 31st.-June 3rd., pp. 614-618, 2022

scientific reports

OPEN

Anti-SARS-CoV-2 IgM/IgG antibodies detection using a patch sensor containing porous microneedles and a paper-based immunoassay

Leilei Bao, Jongho Park, Boyu Qin & Beomjoon Kim✉

Infectious diseases are among the leading causes of mortality worldwide. A new coronavirus named severe acute respiratory syndrome corona virus 2 (SARS-CoV-2) was identified in Wuhan, China in 2019, and the World Health Organization (WHO) declared its outbreak, coronavirus disease 2019 (COVID-19), as a global pandemic in 2020. COVID-19 can spread quickly from person to person. One of the most challenging issues is to identify the infected individuals and prevent potential spread of SARS-CoV-2. Recently, anti-SARS-CoV-2 immunoglobulin M (IgM) and immunoglobulin G (IgG) antibody tests using immunochromatographic methods have been used as a complement to current detection methods and have provided information of the approximate course of COVID-19 infection. However, blood sampling causes pain and poses risks of infection at the needle puncture site. In this study, a novel patch sensor integrating porous microneedles and an immunochromatographic assay (PMNIA) was developed for the rapid detection of anti-SARS-CoV-2 IgM/IgG in dermal interstitial fluid (ISF), which is a rich source of protein biomarkers, such as antibodies. Biodegradable porous microneedles (MNs) made of polylactic acid were fabricated to extract ISF from human skin by capillary effect. The extracted ISF was vertically transported and flowed into the affixed immunoassay biosensor, where specific antibodies could be detected colorimetrically on-site. Anti-SARS-CoV-2 IgM/IgG antibodies were simultaneously detected within 3 min in vitro. Moreover, the limit of detection of anti-SARS-CoV-2 IgM and IgG concentrations was as low as 3 and 7 ng/mL, respectively. The developed device integrating porous MNs and immunochromatographic biosensors is expected to enable minimally invasive, simple, and rapid anti-SARS-CoV-2 IgM/IgG antibody testing. Furthermore, the compact size of the MN and biosensor-integrated device is advantageous for its widespread use. The proposed device has great potential for rapid screening of various infectious diseases in addition to COVID-19 as an effective complementary method with other diagnostic tests.

At the end of 2019, a novel coronavirus, severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), was identified, which causes coronavirus disease 2019 (COVID-19). It spread worldwide within 3 months owing to its high infectivity[1,2]. In March 2020, the World Health Organization (WHO) announced the COVID-19 outbreak as a global pandemic[3]. A COVID-19 infection spreads quickly from person to person and its symptoms include fatigue, cough, fever, dyspnea, anosmia, and ageusia; more severe symptoms include respiratory insufficiency, which can be life-threatening[4,5]. Furthermore, the rate of asymptomatic infections is reported as 16–38%, which brings difficulties in identifying all the individuals with SARS-CoV-2 infected[6]. COVID-19 vaccines are effective in reducing infection risk and virus transmission; however, the proportion of the population fully vaccinated against COVID-19 remains less than 10% in several low-income countries[7,8]. Therefore, one of the current global challenges is to identify both symptomatic and asymptomatic patients as soon as prevent potential spread of SARS-CoV-2.

Currently, real-time reverse transcription polymerase chain reaction (RT-PCR) is the predominant detection method and remains the gold standard for COVID-19 diagnosis[9]. However, there are certain drawbacks

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Biomedical Optics EXPRESS

Localised light delivery on melanoma cells using optical microneedles

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Abstract: Light-based therapy is an emerging treatment for skin cancer, which has received increased attention due to its drug-free and non-invasive approach. However, the limitation of current light therapy methods is the inability for light to penetrate the skin and reach deep lesions. As such, we have developed a polylactic acid (PLA) microneedles array as a novel light transmission platform to perform *in vitro* evaluation regarding the effect of light therapy on skin cancer. For the first time, we designed and fabricated a microneedle array system with a height fixation device that can be installed in a cell culture dish and an LED array for blue light irradiation. The effect of the blue light combined with the microneedles on cell apoptosis was evaluated using B16F10 melanoma cells and analyzed by Hoechst staining. Our results demonstrate that blue light can be transmitted by microneedles to skin cells and effectively affect cell viability.

1. Introduction

Skin cancer is one of the most common forms of cancer, and the number of diagnosed patients increases every year. There are two main types of skin cancer: 1) non-melanoma, which includes squamous cell carcinoma (SCC) and basal cell carcinoma (BSS), and 2) melanoma. Other less common cancer types include atypical fibroxanthoma and skin gland cancer [1–4]. Among them, melanoma is recognized as the most aggressive skin cancer because of its fast proliferation rate and high lethality. Melanoma can be classified into five levels based on the depth of invasions of melanoma cells, where melanoma cells were found from the epidermis until dermis. Therefore, various treatment methods have been proposed, including surgical resection, chemotherapy, radiotherapy, photodynamic therapy, immunotherapy, and targeted therapy to treat melanoma in different levels. However, some ineffective results achieved by most of these treatments are attributed to the high resistance by melanomas. Therefore, the combination strategies, such as a combination of light-based therapy with these treatments has been proposed to improve therapeutic effect for melanoma treatment in early stage [5–7].

In the combination therapies, light- based therapy like phototherapy, also known as light therapy based on photobiomodulation, has been used as an anti-tumor treatment method because of its non-invasive and no drug resistance. Photosensitizers or photon receptors in cells such as flavins, porphyrins, and mitochondria can produce reactive oxygen species (ROS) under specific wavelengths, leading to cell death (apoptosis) [8,9]. In addition, light-emitting diode (LED) therapy has gradually become a novel anti-cancer treatment method because of its large-area irradiation, low cost, and high energy features. In particular, in terms of the effects of different wavelengths, various types of LEDs have been used in the treatment of clinical skin diseases, such as acne, inflammation, and psoriasis [10,11]. Although LED light therapy for melanoma has not

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ORIGINAL ARTICLE

Microfluidic chip connected to porous microneedle array for continuous ISF sampling

Kai Takeuchi[1,2] · Nobuyuki Takama[1] · Kirti Sharma[3,4] · Oliver Paul[3,4] · Patrick Ruther[3,4] · Tadatomo Suga[2] · Beomjoon Kim[1]

Accepted: 8 August 2021

Abstract
Minimally invasive biosensing using microneedles (MNs) is a desirable technology for continuous healthcare monitoring. Among a wide range of MNs, porous MNs are expected to be applied for sampling of interstitial fluids (ISF) by connecting the internal tissue to external measurement devices. In order to realize a continuous measurement of biomarkers in ISF through porous MNs, their integration with a microfluidic chip is a promising approach due to its applicability to micro-total analysis system (μTAS) technology. In this study, we developed a fluidic system to directly interface porous MNs to a microfluidic chip consisting of a capillary pump for the continuous sampling of ISF. The porous and flexible MNs made of PDMS are connected to the microfluidic chip fabricated by standard microelectro-mechanical system (MEMS) processes, showing a continuous flow of phosphate buffered saline (PBS). The developed device will lead to the minimally invasive and continuous biosampling for long-term healthcare monitoring.

Keywords Microneedle · Glucose monitoring · Microchannel · Biosensing

Introduction

Microneedles (MNs) are a rapidly growing field of research and development due to their potential for minimally invasive bioengineering applications. MNs are smaller than conventional hypodermic needles, with lengths of less than 1 mm and widths of less than 500 μm, allowing access to internal tissues without pain or bleeding. Various MNs have been proposed for topical drug delivery and biosensing with functionalized structures and materials implemented as solid, biodissolving, hollow, swelling, or porous MNs.

One of the main targets of the MN-based biosensor is the monitoring of blood glucose concentration. For this, many studies have reported the in vivo and ex vivo measurement of glucose in interstitial fluids (ISF) [1–3]. In addition to the glucose measurement for point-of-care usage, continuous glucose monitoring (CGM) is also targeted as an effective approach to prevent, diagnose, and treat diabetes. From the perspective of CGM, in vivo glucose monitoring has been successfully demonstrated using solid MNs as electrodes for electrochemical measurements [4–6]. However, the MN-based in vivo microelectrodes face challenges such as surface fouling and the biological risk of MN breakage in the case of their long-term usage.

Compared to solid MNs, porous MNs are more suitable for biosampling using capillary action [7–10]. The porous structure absorbs ISF through interconnected pores in the MN body, but has relatively poor mechanical and geometric properties. Porous MNs with sharp tips are more likely to break during insertion due to their low mechanical reliability.

To overcome these drawbacks, we proposed a porous and flexible MN array made of dimethylpolysiloxane (PDMS) [11]. The porous PDMS MNs have a “sponge-like” structure, which allows high durability against mechanical stresses during long-term use and ISF extraction at the same time. In our experiments, the porous PDMS MNs were able

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Drug Delivery and Translational Research
https://doi.org/10.1007/s13346-021-01045-x

ORIGINAL ARTICLE

Recent advances in porous microneedles: materials, fabrication, and transdermal applications

Leilei Bao[1] · Jongho Park[1] · Gwenaël Bonfante[2] · Beomjoon Kim[1,2]

Accepted: 8 August 2021

Abstract
In the past two decades, microneedles (MNs), as a painless and simple drug delivery system, have received increasing attention for various biomedical applications such as transdermal drug delivery, interstitial fluid (ISF) extraction, and biosensing. Among the various types of MNs, porous MNs have been recently researched owing to their distinctive and unique characteristics, where porous structures inside MNs with continuous nano- or micro-sized pores can transport drugs or biofluids by capillary action. In addition, a wide range of materials, including non-polymers and polymers, were researched and used to form the porous structures of porous MNs. Adjustable porosity by different fabrication methods enables the achievement of sufficient mechanical strength by optimising fluid flows inside MNs. Moreover, biocompatible porous MNs integrated with biosensors can offer portable detection and rapid measurement of biomarkers in a minimally invasive manner. This review focuses on several aspects of current porous MN technology, including material selection, fabrication processes, biomedical applications, primarily covering transdermal drug delivery, ISF extraction, and biosensing, along with future prospects as well as challenges.

Introduction

Microneedles (MNs), having microscopic needle structures, were initially developed to facilitate transdermal drug delivery by piercing human skin and providing transport conduits across the stratum corneum with a thickness of 10–15 µm [1, 2]. MNs have received significant attention in recent years owing to their micro-sized structure that can penetrate the skin painlessly without stimulating nerve endings, which results in minimal invasiveness and better patient compliance [3–5]. Taking advantage of its microscopic structures, MNs have been developed as a suitable tool for painless drug delivery and biosensing. According to the matrix material and structure, MNs are divided into six categories, as depicted in Fig. 1: solid, coated, hollow, dissoluble, swellable, and porous MNs.

The first MNs, which aimed at enhancing the skin permeability for transdermal drug delivery, were of the solid type [1]. Solid MNs are generally fabricated using silicon, metal, or polymer materials, which have sufficient mechanical strength to puncture the stratum corneum and penetrate the epidermis of human skin [6–11]. After the penetration and removal of solid MNs from the skin, drug components are transported into the dermis layer through pathways formed by the MNs. Because of these pathways, the skin permeability can be enhanced to facilitate transdermal delivery of both micro- and macro-molecular (over 600 Da) drug material. However, accurate control of the drug amount is difficult because of the gradual closure of formed pores over time. Moreover, the leftovers of broken solid MNs, which are manufactured from metals, silicon, and non-biocompatible polymers, are considered harmful to the human body because they cannot be biodegraded.

Conversely, the development of coated MNs overcame the issue of pore closure after the penetration and removal of solid MNs. By coating the drug material on the surface of the MNs, the drug can diffuse into the skin directly while maintaining the pathways formed by the penetration without leaving drug waste on the surface of the skin. Similar to the solid type, the coated MNs are fabricated using silicon, metals, or polymers [12–14]. Coated MNs have the advantages of achieving controllable drug dose by adjusting the amount of coated drug. In addition, drug stability can be

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ARTICLE Open Access

Optimization of the fused deposition modeling-based fabrication process for polylactic acid microneedles

Libo Wu[1], Jongho Park[1], Yuto Kamaki[1] and Beomjoon Kim[1]

Abstract

A microneedle (MN) array is a novel biomedical device adopted in medical applications to pierce through the stratum corneum while targeting the viable epidermis and dermis layers of the skin. Owing to their micron-scale dimensions, MNs can minimize stimulations of the sensory nerve fibers in the dermis layer. For medical applications, such as wound healing, biosensing, and drug delivery, the structure of MNs significantly influences their mechanical properties. Among the various microfabrication methods for MNs, fused deposition modeling (FDM), a commercial 3D printing method, shows potential in terms of the biocompatibility of the printed material (polylactic acid (PLA)) and preprogrammable arbitrary shapes. Owing to the current limitations of FDM printer resolution, conventional micron-scale MN structures cannot be fabricated without a post-fabrication process. Hydrolysis in an alkaline solution is a feasible approach for reducing the size of PLA needles printed via FDM. Moreover, weak bonding between PLA layers during additive manufacturing triggers the detachment of PLA needles before etching to the expected sizes. Furthermore, various parameters for the fabrication of PLA MNs with FDM have yet to be sufficiently optimized. In this study, the thermal parameters of the FDM printing process, including the nozzle and printing stage temperatures, were investigated to bolster the interfacial bonding between PLA layers. Reinforced bonding was demonstrated to address the detachment challenges faced by PLA MNs during the chemical etching process. Furthermore, chemical etching parameters, including the etchant concentration, environmental temperature, and stirring speed of the etchant, were studied to determine the optimal etching ratio. To develop a universal methodology for the batch fabrication of biodegradable MNs, this study is expected to optimize the conditions of the FDM-based fabrication process. Additive manufacturing was employed to produce MNs with preprogrammed structures. Inclined MNs were successfully fabricated by FDM printing with chemical etching. This geometrical structure can be adopted to enhance adhesion to the skin layer. Our study provides a useful method for fabricating MN structures for various biomedical applications.

Introduction

A commercially available microneedle (MN) patch is primarily a cosmetic product that contains micron-sized polymer needles. The concept of MNs was first introduced in 1976 for transdermal drug delivery[1]. Biomedical applications of MNs have been further developed for wound healing and biosensing[2–4]. MNs can bypass the stratum corneum, which is the outermost barrier layer of the skin. Active pharmaceutical ingredients (APIs) are subsequently administered to the viable epidermis and dermis layers of the skin. MN-mediated applications enable delivered APIs to block abnormal cell cycles in the skin, which can treat lesions[5]. Compared with the systemic drug delivery method, drug delivery by MNs targeting affected dermal cells can improve the efficiency of

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These authors contributed equally: Libo Wu, Jongho Park, Yuto Kamaki, Beomjoon Kim

TAKAMIYA Laboratory

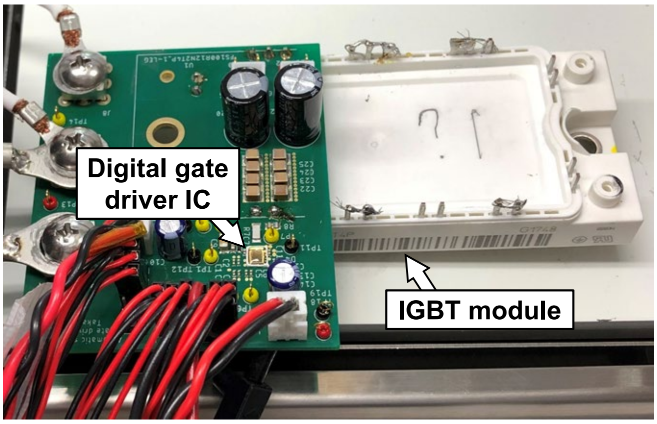
Current Research Activities 2022-2023

Takamiya Laboratory

1. Research Topics

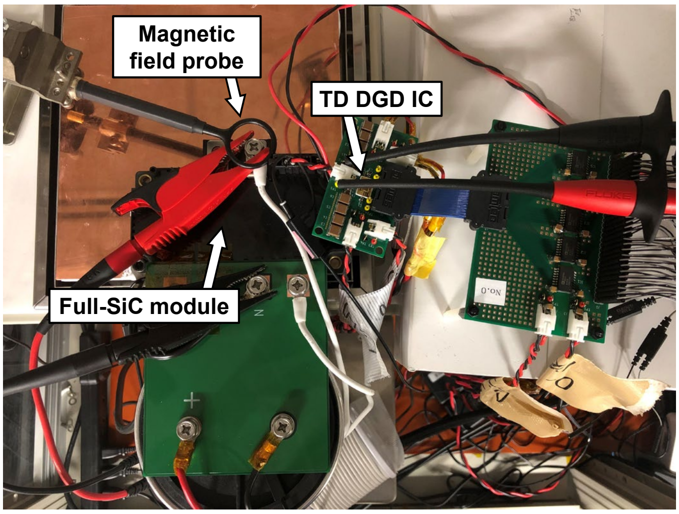
1.1 Digital Gate Driver IC with Fully Integrated Automatic Timing Control Function in Stop-and-Go Gate Drive for IGBTs

To reduce loss and noise during switching of IGBTs at low cost even under changing operating conditions, namely load current (I_L) and junction temperature (T_J) variation, the world's first active digital gate driver IC with fully integrated automatic timing control function that achieves both single-chip integration and real-time control is developed. In order to handle various IGBTs with one type of gate driver, the proposed IC also has the world's first 6-bit programmable gate current function in the closed-loop active gate drivers. In the double pulse test of IGBT, compared with the conventional single-step gate drive, the stop-and-go gate drive with automatic timing control using the proposed IC effectively reduces the switching loss (E_{LOSS}) under the collector current overshoot ($I_{OVERSHOOT}$)-aligned condition and reduces $I_{OVERSHOOT}$ under E_{LOSS}-aligned condition with I_L ranging from 10 A to 80 A and T_J ranging from 25 ˚C (room temperature) to 125 ˚C, and particularly at the condition of I_L = 80 A at 25 ˚C, the proposed gate driver reduces E_{LOSS} by 49 % under the $I_{OVERSHOOT}$-aligned condition and reduces $I_{OVERSHOOT}$ by 33 % under the E_{LOSS}-aligned condition.



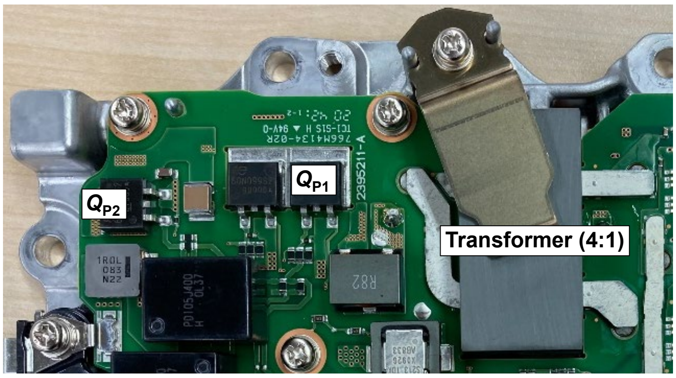
1.2 Sub-0.5 ns Step, 10-bit Time Domain Digital Gate Driver IC for Reducing Radiated EMI and Switching Loss of SiC MOSFETs

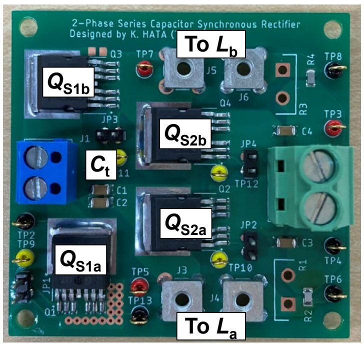
A sub-0.5 ns step, 10-bit time domain digital gate driver (TD DGD) IC is proposed for SiC MOSFETs to reduce both the radiated EMI and the switching loss (E_{LOSS}). Unlike the conventional current-domain DGDs, the proposed TD DGD has the advantage that the gate current is binary and only one period is digitally changed, making it easy to search for the optimal gate waveform that reduces both EMI and E_{LOSS}. Using TD DGD IC fabricated with 180-nm BCD process, the radiated EMI spectrums from 30 MHz to 100 MHz and E_{LOSS} are measured in the double pulse test of a full SiC module at 600 V and 300 A. The proposed active gate drive using TD DGD IC reduces E_{LOSS} by 50 % and 39 % compared with the conventional single-step gate drive while satisfying an EMI limit in turn-on and turn-off, respectively.



1.3 <u>2-Phase Series Capacitor Synchronous Rectifier in Active Clamp Forward Converter</u>

A 2-phase series capacitor synchronous rectifier (SC-SR) in active clamp forward (ACF) converters is proposed to solve the inductor cooling problems caused by the recent trend of increasing the output current. The proposed 2-phase SC-SR can achieve the interleaved operation by adding only one flying capacitor to the 2-parallel conventional SRs without increasing the number of the primary circuit elements and transformer. Furthermore, the proposed 2-phase SC-SR can achieve the automatic inductor current balancing, which helps distribute the heat evenly in the two inductors. In the measurement at 140 V-to-5 V conversion, the peak efficiency of the ACF converters with the proposed 2-phase SC-SR and conventional SR was 90.3 % and 85.9 % at 28 A_{OUT}, respectively, resulting in the improvement in efficiency by 4.4 %. In addition, the interleaved operation of the proposed 2-phase SC-SR reduced the output current ripple from 10.8 A to 6.4 A compared to the conventional SR at 40 A_{OUT}. The current imbalance between the two output inductors of the proposed 2-phase SC-SR was less than 10% under heavy load even without any control or compensation, demonstrating the practicability of the proposed 2-phase SC-SR in ACF converters.





2. Research Achievements

2.1 Number of original journal papers: 3

2.2 International conference: 7 (including 0 invited presentations)

2.3 Domestic conference: 13(including 2 invited presentations)

2.4 Number of patents: 3

3. List of awards

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4. Research Grants

4.1 Total number of research grants: 5

4.2 Number of collaboration research with industries: 6

4.3 List of major research grants (serving as Principal Investigator)

- Research Grant for “ Grant-in-Aid for Exploratory Research” from KAKENHI

5. Education

5.1 Number of Ph.D. students (including current students): 1

5.2 Number of master students (including current students):4

5.3 Number of other students: 1

6. Publication list

Journal Papers

1. Z. Lou, T. Mamee, K. Hata, M. Takamiya, S. -I. Nishizawa, and W. Saito, "IGBT Power Module Design for Suppressing Gate Voltage Spike at Digital Gate Control," IEEE Access, Vol.11, pp. 6632 - 6640, Jan. 2023.
2. H. Qiu, T. Sakurai, and M. Takamiya, "A 6.78-MHz Multiple-Transmitter Wireless Power Transfer System With Efficiency Maximization by Adaptive Magnetic Field Adder IC," IEEE Journal of Solid-State Circuits, Vol.57, No.8, pp. 2390 - 2403, Aug. 2022.
3. Y. S. Cheng, D. Yamaguchi, T. Mannen, K. Wada, T. Sai, K. Miyazaki, M. Takamiya, and T. Sakurai, "High-Speed Searching of Optimum Switching Pattern for Digital Active Gate Drive to Adapt to Various Load Conditions," IEEE Transactions on Industry Electronics, Vol.69, No.5, pp. 5185 - 5184, May 2022.

Conference Presentations

C1. D. Zhang, K. Horii, K. Hata, and M. Takamiya, "Digital Gate Driver IC with Fully Integrated Automatic Timing Control Function in Stop-and-Go Gate Drive for IGBTs," IEEE Applied Power Electronics Conference and Exposition (APEC), Orlando, USA, pp. 1225-1231, March 2023.

C2. K. Hata, S. Suzuki, K. Watanabe, K. Nagayoshi, and M. Takamiya, "2-Phase Series Capacitor Synchronous Rectifier in Active Clamp Forward Converter," IEEE Applied Power Electronics Conference and Exposition (APEC), Orlando, USA, pp. 906-911, March 2023.

C3. T. Inuma, K. Hata, T. Sai, W. Saito, and M. Takamiya, "Two Stop-and-Go Gate Driving to Reduce Switching Loss and Switching Noise in Automotive IGBT Modules," IEEE Southern Power Electronics Conference (SPEC), Nadi, Fiji, pp. 1-7, Dec. 2022.

C4. H. Zhang, H. Yamasaki, K. Hata, I. Omura, and M. Takamiya, "Overcurrent Detection Method by Monitoring Gate Voltage While Periodically Repeating Discharging and Charging of Constant Gate Charge in IGBTs," IEEE Southern Power Electronics Conference (SPEC), Nadi, Fiji, pp. 1-5, Dec. 2022.

C5. K. Horii, R. Morikawa, K. Hata, K. Morokuma, Y. Wada, Y. Obiraki, Y. Mukunoki, and M.

Takamiya, "Sub-0.5 ns Step, 10-bit Time Domain Digital Gate Driver IC for Reducing Radiated EMI and Switching Loss of SiC MOSFETs," IEEE Energy Conversion Congress & Exposition (ECCE), Detroit, USA, pp. 1-8, Oct. 2022.

C6. H. Yamasaki, K. Hata, and M. Takamiya, "Estimation of Both Junction Temperature and Load Current of IGBTs from Output Voltage of Gate Driver," International Power Electronics Conference (IPEC-Himeji 2022 -ECCE Asia-), Himeji, Japan, pp. 453-460, May 2022.

C7. K. Horii, K. Hata, R. Wang, W. Saito, and M. Takamiya, "Large Current Output Digital Gate Driver Using Half-Bridge Digital-to-Analog Converter IC and Two Power MOSFETs," IEEE International Symposium on Power Semiconductor Devices and ICs (ISPSD), Vancouver, Canada, pp. 293 – 296, May 2022.

2-Phase Series Capacitor Synchronous Rectifier in Active Clamp Forward Converter

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***Abstract*— A 2-phase series capacitor synchronous rectifier (SC-SR) in active clamp forward (ACF) converters is proposed to solve the inductor cooling problems caused by the recent trend of increasing the output current. The proposed 2-phase SC-SR can achieve the interleaved operation by adding only one flying capacitor to the 2-parallel conventional SRs without increasing the number of the primary circuit elements and transformer. Furthermore, the proposed 2-phase SC-SR can achieve the automatic inductor current balancing, which helps distribute the heat evenly in the two inductors. In the measurement at 140 V-to-5 V conversion, the peak efficiency of the ACF converters with the proposed 2-phase SC-SR and conventional SR was 90.3 % and 85.9 % at 28 AOUT, respectively, resulting in the improvement in efficiency by 4.4 %. In addition, the interleaved operation of the proposed 2-phase SC-SR reduced the output current ripple from 10.8 A to 6.4 A compared to the conventional SR at 40 AOUT. The current imbalance between the two output inductors of the proposed 2-phase SC-SR was less than 10% under heavy load even without any control or compensation, demonstrating the practicability of the proposed 2-phase SC-SR in ACF converters.**

Keywords*— *Active clamp forward converter, Synchronous rectifier, Series capacitor converter, current sharing.

I. Introduction

Active clamp forward (ACF) converters [1–2] are suitable for a high step-down conversion in applications such as EV/HEV, servers, and data centers [3–5]. The trend of increasing the output current in recent years, however, causes problems due to the increased heat generation in the output inductor of the ACF converter with the conventional synchronous rectifier (SR), which is shown in Fig. 1. To suppress the heat generated in the inductor, it is conceivable to connect the converters in parallel and perform the interleaved operation [6–8]. Alternatively, to reduce the increase in the number of elements including transformers, paralleling only the secondary circuit is also possible, but this way cannot achieve interleaving because the transformer is shared and the secondary circuit operation depends on the secondary current of the transformer. In addition, paralleling two inductors without interleaving increases the output current and voltage ripple due to the reduced equivalent inductance. In order to prevent this, it is necessary to increase the inductor size.

To solve these problems, this paper proposes a 2-phase series capacitor synchronous rectifier (SC-SR), which can be regarded as applying a circuit topology of non-isolated 2-phase series capacitor (SC) buck converters with two output inductors [9–13] to the synchronous rectifier in isolated DC-DC converters. Similar to the non-isolated SC buck converter, the proposed SC-SR can achieve the interleaved operation and current balancing of the two output inductors in the isolated DC-DC converter, distributing the heat evenly to the two inductors. In this paper, the operating principle of the proposed 2-phase SC-SR in an ACF converter is presented and the practical feasibility of the proposed 2-phase SC-SR is demonstrated by experiments.

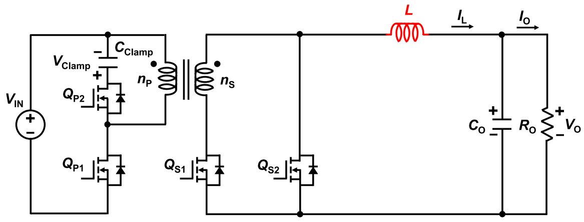

Fig. 1. Active clamp forward converter with conventional synchronous rectifier.

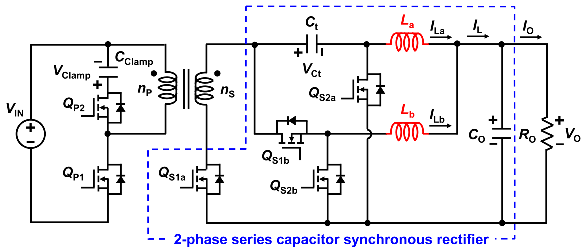

Fig. 2. Active clamp forward converter with proposed 2-phase series capacitor synchronous rectifier (SC-SR).

II. Proposed 2-Phase Series Capacitor Synchronous Rectifier (SC-SR)

A. Circuit operation

Fig. 2 shows the ACF converter with the proposed 2-phase SC-SR, which consists of 4 switches, 2 inductors, and 1 flying capacitor. The phase A consists of Q_{S1a} and Q_{S2a}, where Q_{S1a} is operated synchronously with Q_{P1}, and Q_{S2a} is alternately turned on and off with Q_{S1a} in continuous current mode. The phase B is composed of Q_{S1b} and Q_{S2b}, where Q_{S1b} is operated with 180° phase difference from Q_{S1a} for interleaving and Q_{S2b} are also alternately turned on and off Q_{S1b} in continuous current mode. The duty cycles D_a and D_b are defined as the on-time ratios of Q_{S1a} and Q_{S1b} in each phase and this paper assumes that D_a and D_b do not exceed 50 %.

Focusing on the circuit operation in a steady state, the main circuit states and ideal waveforms of the proposed 2-phase SC-SR are shown in Figs. 3 and 4. During state 1, Q_{S1a} in the phase A is turned on synchronously with Q_{P1}, where the series capacitor C_t is connected in series with L_a and the transformer supplies the current I_{La}, which charges C_t. Also, Q_{S2b} is on in the phase B, so I_{Lb} freewheels through Q_{S2b}. During state 2, Q_{S1a} is turned off synchronously with Q_{P1} and the transformer stops supplying the current, so the current path of I_{La} changes from Q_{S1a} to Q_{S2a}. Since I_{Lb} continues to freewheel through Q_{S2b}, C_t is neither charged nor discharged, keeping the voltage V_{Ct} constant. During state 3, Q_{S1b} turns on instead of Q_{S2b} in the phase B, where C_t is connected in series with L_b and supplies the current I_{Lb}, which discharges C_t. Also, Q_{S2a} is on in the phase A and I_{La} continues to flow through Q_{S2a}. Finally,

A 6.78-MHz Multiple-Transmitter Wireless Power Transfer System With Efficiency Maximization by Adaptive Magnetic Field Adder IC

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***Abstract*—A 6.78-MHz multiple-transmitter (TX) wireless power transfer (WPT) system was presented. An adaptive magnetic field adder (AMFA) IC was proposed, for the first time, to enable the maximization of the system efficiency (η_{SYS}) by adaptively optimizing the amplitude and phase of the current in each TX coil on the basis of the coupling coefficient (k) between each TX coil and the receiver (RX) coil. Under the optimal condition, the current in each TX coil is proportional to k between the TX coil and the RX coil. For the independent control of the current in each TX coil, a selectively activated shared-half-bridge (HB) power amplifier (PA) together with an alternate TX coil array was proposed. To sense a small k, duty-ratio control was proposed in the integrated k sensor. The AMFA IC was fabricated by a 0.18-μm CMOS process with 1.8-V devices. The peak power conversion efficiency of the proposed PA reached 74%. The k sensor could accurately measure k with a percentage error within ±2.5%. A WPT system consisting of a 4×4 TX coil array driven by four AMFA ICs and a single RX coil was implemented. Experimental results showed that, compared with the conventional system, η_{SYS} was increased from 0.11% to 51% with a load power of 576 mW when the RX coil was perpendicular to the TX coils. When the RX coil was parallel to the TX coils, a η_{SYS} of 63%, which is higher than those in previous works, was also achieved.**

***Index Terms*—Adaptive magnetic field adder (AMFA), alternate coil array, amplitude and phase control, coupling coefficient sensor, double-off control, duty-ratio control, efficiency maximization, magnetic resonance coupling, multiple transmitter (TX) coils, near field, power amplifier (PA) topology for multiple TX coils, shared topology, wireless power transfer (WPT).**

I. Introduction

WIRELESS power transfer (WPT) based on magnetic resonance coupling is a promising technology for the charging of mobile devices, including mobile phones, smart watches, and so on [1]. Instead of inserting the charging cable and leaving the devices in a corner every time, we would be able to operate them and move freely if they could be wirelessly charged; this would greatly improve the user experience.

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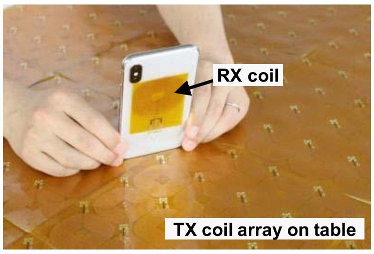

Fig. 1. WPT scenario where a mobile device with the RX coil is wirelessly charged by the TX coil array placed on a table.

To realize this vision where the receiver (RX) can change its position and sometimes be perpendicular to the transmitter (TX), many related works have been published. In a WPT system, the most important target has been the improvement of its efficiency and robustness against the variation in the position of the RX relative to the TX.

In a system using a single TX coil, the power transferred through the magnetic field is highly directional and its value drops very rapidly with distance and misalignment between the TX and RX coils. As a result, it is always required that the RX coil be perfectly aligned with the TX coil, which degrades the user experience. To alleviate this problem, several methods, such as coil design [2]–[4], new compensation topologies [5]–[7], new circuit topologies [8]–[12], and impedance matching techniques [13]–[16], have been proposed. However, when the coupling coefficient (k) between the TX and RX coils is small, the benefits that these methods yield can be limited. Under an extreme condition where k is zero, no power can be transferred to the RX coil. Physically, this case corresponds to the RX coil being perpendicular to the TX coil with the center of the TX coil being on the plane of the RX coil [17].

Rather than using a single TX coil, the system in which multiple TX coils [18]–[27] are used has gathered much attention, owing to its increased design freedom. Fig. 1 shows an application scenario in which a mobile phone is charged using multiple TX coils on a table. The conventional methods include selectively driving one of the multiple TX coils [18], [19], as shown in Fig. 2(a), and driving all TX coils with the same current [20], as shown in Fig. 2(b). On the other hand, as shown in Fig. 2(c), with the optimal control of the amplitude and phase of the current in each TX coil, the magnetic fields from multiple TX coils can be constructively added at the position of the RX coil to maximize the system

Digital Gate Driver IC with Fully Integrated Automatic Timing Control Function in Stop-and-Go Gate Drive for IGBTs

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***Abstract*— To reduce loss and noise during switching of IGBTs at low cost even under changing operating conditions, namely load current (I_L) and junction temperature (T_J) variation, the world's first active digital gate driver IC with fully integrated automatic timing control function that achieves both single-chip integration and real-time control is developed. In order to handle various IGBTs with one type of gate driver, the proposed IC also has the world's first 6-bit programmable gate current function in the closed-loop active gate drivers. In the double pulse test of IGBT, compared with the conventional single-step gate drive, the stop-and-go gate drive with automatic timing control using the proposed IC effectively reduces the switching loss (E_{LOSS}) under the collector current overshoot ($I_{OVERSHOOT}$)-aligned condition and reduces $I_{OVERSHOOT}$ under E_{LOSS}-aligned condition with I_L ranging from 10 A to 80 A and T_J ranging from 25 ˚C (room temperature) to 125 ˚C, and particularly at the condition of I_L = 80 A at 25 ˚C, the proposed gate driver reduces E_{LOSS} by 49 % under the $I_{OVERSHOOT}$-aligned condition and reduces $I_{OVERSHOOT}$ by 33 % under the E_{LOSS}-aligned condition.**

***Keywords*— IGBT, IC, surge current, energy loss, active gate driver**

I. Introduction

A lot of active gate drivers (AGDs), where the gate driving waveform is controlled during the turn-on/off transients, have been proposed to reduce both the switching loss and the switching noise of power devices. AGDs can be classified into two types, open-loop control [1-6] and closed-loop control [7-19, 22]. The closed-loop AGDs are required instead of the open-loop AGDs, because the optimal driving waveform changes depending on the operating conditions (e.g. load current and temperature) [20]. Fig. 1 summarizes the design choices in conventional closed-loop AGDs. To make the closed-loop AGDs practical, the following three points are required: (1) single-chip integration instead of PCB implementation for lower cost, (2) real-time control instead of iterative control to reliably handle dynamic change of operating conditions, and (3) programmable AGDs instead of fixed-function AGDs that require individual optimization for different product variety of power devices. In the closed-loop

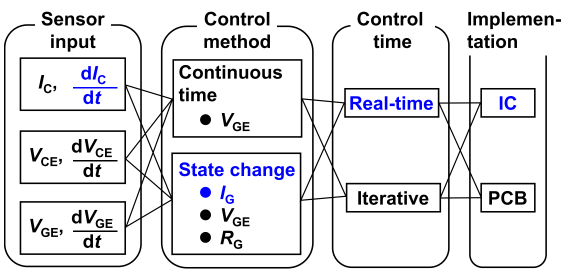

Fig. 1: Design choices in closed-loop AGDs. This work is shown in blue.

AGDs, however, no previous paper has realized (1) and (2) simultaneously, and no previous paper on (3) has been published. To solve the problems, in this paper, a digital gate driver (DGD) IC with fully integrated automatic timing control (ATC) function for IGBTs that realizes all of (1) to (3) is proposed. The design choices in this paper are shown in blue in Fig. 1.

II. Proposed Digital Gate Driver IC with Automatic Timing Control

Figs. 2 and 3 show a circuit schematic and a timing chart of the proposed DGD IC with ATC, respectively. In the following, turn-on is discussed, whereas the exact same is true for turn-off. The IC includes dI_C/dt detector for the state change, controller for ATC, and a 6-bit current-source type

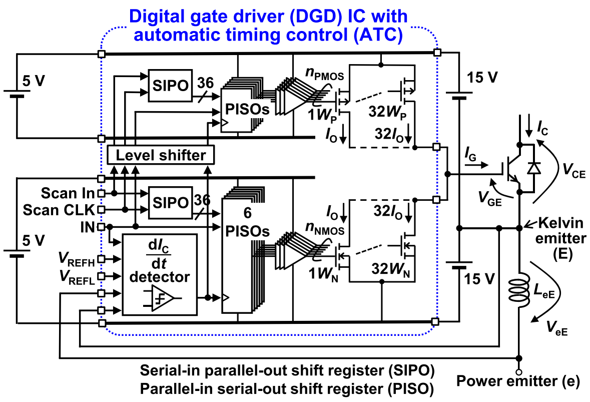

Fig. 2: Circuit schematic of proposed DGD IC with ATC.

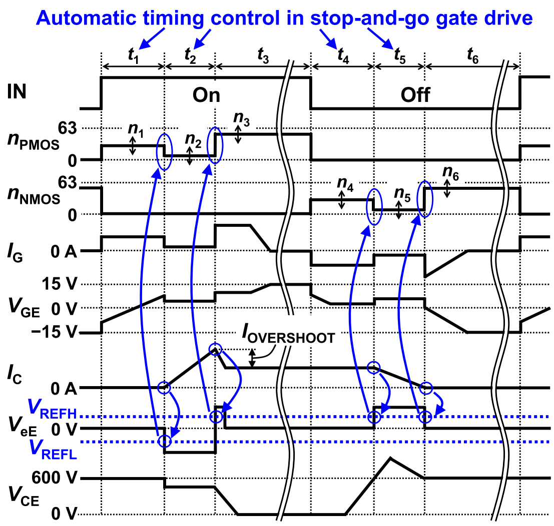

Fig. 3: Timing chart of proposed DGD IC with ATC.

Large Current Output Digital Gate Driver Using Half-Bridge Digital-to-Analog Converter IC and Two Power MOSFETs

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***Abstract*—An 8-bit digital gate driver (DGD) using a half-bridge digital-to-analog converter (HB DAC) IC and two power MOSFETs is proposed to enable the output voltage swing of ± 15 V and the large gate current up to 58 A for a 6500 V, 1000 A IGBT module. In the turn-on measurements of IGBT at 3000 V and 1000 A, compared with the conventional single-step gate driving, the proposed active gate driving using DGD reduces the switching loss from 6.9 J to 4.8 J by 30 % at the same current overshoot of 1.3 kA and reduces the current overshoot from 1560 A to 1330 A by 15 % at the same switching loss of 5 J, which clearly shows the advantage of DGD for the 6500 V, 1000 A IGBT module. This paper is the first to demonstrate the advantages of DGD in the high-voltage, large-current IGBT modules.**

***Keywords*—gate driver, IGBT, switching loss, high voltage**

I. Introduction

High-voltage, large-current IGBT modules (e.g. ratings of 6500 V, 1000 A) are used in many social infrastructure fields including high-voltage DC transmission systems and train traction systems [1]. In addition to improving IGBTs themselves, gate driving technologies can be used to reduce the loss of IGBTs. Recently, many papers have been published on the simultaneous reduction of both switching loss (E_{LOSS}) and switching noise by active gate waveform control using digital gate drivers (DGDs) [2-8]. Conventional DGDs, however, are difficult to apply to the 6500 V, 1000 A IGBT modules, because the modules require DGD with (1) the output voltage swing (V_{SWING}) of ± 15 V to prevent a false turn-on and (2) the gate current (I_G) of up to around 20 A because of the large gate capacitance. For example, V_{SWING} is 3.3 V [3], 5 V [4–5, 7], 15 V [2], 18 V [6] and 20 V [8], and the maximum I_G is between 5 A [2, 4] and 42 A [5].

To solve the problems, in this paper, an 8-bit DGD using a half-bridge digital-to-analog converter (HB DAC) IC and two power MOSFETs is proposed to enable V_{SWING} of ± 15 V and large I_G up to 58 A for the 6500 V, 1000 A IGBT modules. 30 % reduction of E_{LOSS} and 15 % reduction of the current overshoot ($I_{OVERSHOOT}$) compared with the conventional single-step gate driving in the turn-on of the 6500 V, 1000 A IGBT module at 3000 V and 1000 A are experimentally shown. This paper is the first to demonstrate the advantages of DGD in the high-voltage, large-current IGBT modules.

II. Design of Digital Gate Driver Using HB DAC IC and Two Power MOSFETs

Figs. 1 to 3 show a circuit schematic of the proposed DGD including HB DAC IC and two power MOSFETs (Q_1 and Q_2 : BSC094N06LS5, 60 V, 47 A), a block diagram of the proposed HB DAC IC, and a timing chart of DGD, respectively. DGD is a current-source gate driver. The novelty of this work is that power MOSFETs are used as the output stage of the gate driver to achieve large I_G, and DGD operation is achieved by digitally controlling the gate amplitude (V_{GSH} and V_{GSL}) of the power MOSFETs operating in the saturation region instead of the linear region using the proposed HB DAC IC to achieve the current-source gate driver. As shown in Fig. 2, HB DAC IC includes two DACs operating with different power supply rails, shift registers for serial inputs to reduce the number of input pins, and an edge detector to generate pulse signals from externally supplied "Timing" signal. HB DAC IC does not include the driver transistors. If all the functions are integrated into a single IC, the chip size will be huge and the cost will be high. By controlling the gate voltage of Q_1 (V_{GSH}) with a 16-bit input DAC (Fig. 2), I_G can be digitally varied four times at turn-on (Fig. 3). The four periods from t_1 to t_4 are determined by "Timing" signal, and t_1 to t_4 can be changed independently. The same is true for turn-off.

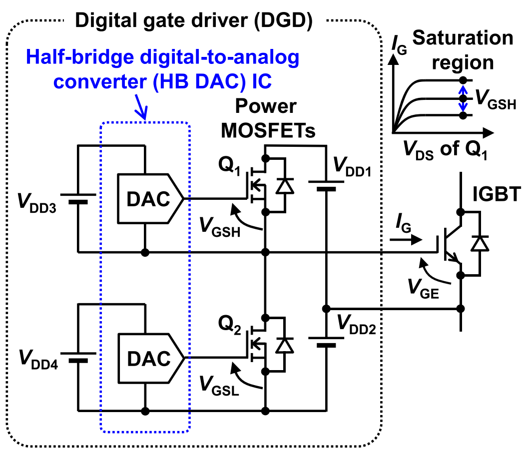

Fig. 1. Circuit schematic of proposed digital gate driver (DGD) including HB DAC IC and two power MOSFETs.

Sub-0.5 ns Step, 10-bit Time Domain Digital Gate Driver IC for Reducing Radiated EMI and Switching Loss of SiC MOSFETs

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***Abstract*— A sub-0.5 ns step, 10-bit time domain digital gate driver (TD DGD) IC is proposed for SiC MOSFETs to reduce both the radiated EMI and the switching loss (E_{LOSS}). Unlike the conventional current-domain DGDs, the proposed TD DGD has the advantage that the gate current is binary and only one period is digitally changed, making it easy to search for the optimal gate waveform that reduces both EMI and E_{LOSS}. Using TD DGD IC fabricated with 180-nm BCD process, the radiated EMI spectrums from 30 MHz to 100 MHz and E_{LOSS} are measured in the double pulse test of a full SiC module at 600 V and 300 A. The proposed active gate drive using TD DGD IC reduces E_{LOSS} by 50 % and 39 % compared with the conventional single-step gate drive while satisfying an EMI limit in turn-on and turn-off, respectively.**

***Keywords*—EMI, switching loss, gate driver, time domain**

I. Introduction

Fast switching of SiC MOSFETs reduces the switching loss (E_{LOSS}), while the radiated EMI increases. Active gate driver [1–2] and digital gate driver (DGD) [3–8], which digitally controls the gate driving current of power devices during turn-on/off transients, are promising technologies to overcome the trade-off between EMI and E_{LOSS}. Addressing the radiated EMI using conventional DGDs, however, has problems.

Two methods of implementing DGD can be considered, a current-domain (CD) DGD and a time domain (TD) DGD. Fig. 1 (a) shows CD DGD, where the time step (Δt) is fixed and the gate current (I_G) is digitally n-bit controlled. All conventional DGDs [3–8] are CD DGDs. Fig. 1 (b) shows TD DGD, where I_G is binary and each time step is digitally n-bit controlled. Since the radiated EMI is generated at the moment when the SiC MOSFET switches, in order to control the radiated EMI using DGD, it is necessary to change I_G using DGD precisely targeting the moment of the switching. When the frequency range of the radiated EMI is from 30 MHz to 1 GHz, the range of time steps required for DGD is from 0.5 ns (= 1 / 1 GHz / 2) to 17 ns (= 1 / 30 MHz / 2). In the conventional CD DGDs, however, when Δt is reduced below 17 ns to control the radiated EMI, the number of steps (m) increases and the number of DGD parameter combinations explodes to $(2^n)^m$ as shown in Fig. 1 (a). For example, in DGD with n = 14 and m = 104 developed for

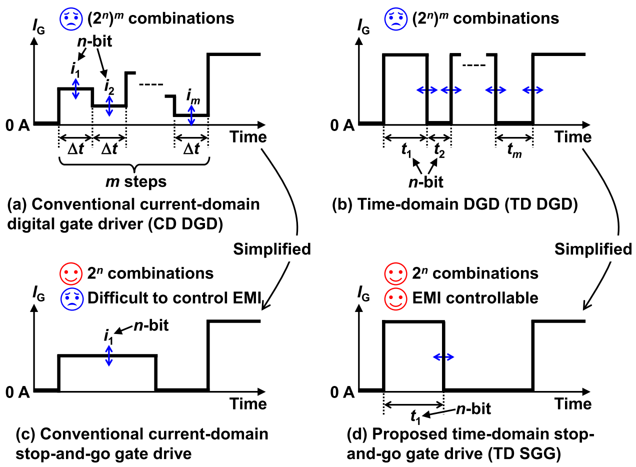


Fig. 1. Gate current (I_G) waveforms in digital gate drivers.

the radiated EMI in GaN [8], it is extremely difficult to find the optimal DGD parameters, because the number of DGD parameter combinations is $(2^{14})^{104}$. Therefore, in order to control the radiated EMI with reducing the number of DGD parameter combinations, TD DGD is better than CD DGD to change I_G precisely targeting the moment of the switching.

In addition, in order to reduce the number of DGD parameter combinations from $(2^n)^m$ to 2^n, a current-domain stop-and-go gate drive shown in Fig. 1 (c) is reported in [9, 10], while this is not suitable for the radiated EMI control, because the timing control is not possible. To solve the problems, a time-domain stop-and-go gate drive (TD SGG) shown in Fig. 1 (d) is proposed in this paper. By digitally controlling only t_1 in 10-bit, sub-0.5 ns steps, the proposed TD SGG reduces E_{LOSS} by 50 % and 39 % compared with the conventional single-step gate drive (SSG) while satisfying an EMI limit in turn-on and turn-off, respectively.

MIZOGUCHI Laboratory

Current Research Activities 2022-2023

Mizoguchi Laboratory

1. Research Topics

1.1 Application of Machine Learning for Spectrum Analysis: Tree Structure based Unsupervised and Supervised Learning and Constructing Spectrum Database

Characterization of atomic and electronic structures is indispensable in materials chemistry. The process is necessary for the development of new materials and a variety of analytical techniques are employed based on the relevant system, the desired information, and the required resolution. Chemical reactions particularly occur in very local environments and this makes an understanding of the atomic and electronic structures very important. Moreover, during chemical reactions, the atomic and electronic structures of molecules undergo dynamic changes depending on the reaction environment, and an understanding of the entire transition is also imperative.

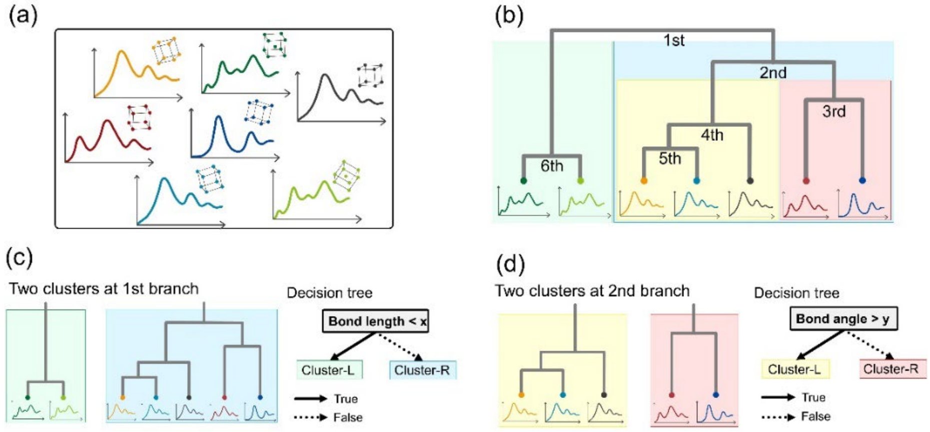
In the context of the foregoing, core-loss spectroscopy is a promising technique for the analysis of both atomic and electronic structures with nano-order spatial resolution and time resolution. The technique utilizes X-ray or fast electrons as probes, and the spectral data thus reflects the atomic and electronic structures at the nano- subnano-scale. It specifically offers very local information such as the constituent elements, oxidation state, coordination environment, and chemical bonds. Furthermore, core-loss experiments can be performed in operando, in the case of which the spectrum changes also reflect the atomic and electronic structure transitions that occur during a chemical reaction. This has informed the broad use of core-loss spectroscopic techniques in the investigation of chemical materials science processes such as catalysis, oxygen reduction reaction (ORR), and electrochemical reaction.

Machine learning (ML) techniques have been recently widely utilized in chemical materials science. They reduce the computation demand by replacing theoretical calculation with predictors that can estimate material functions with very low costs. Using such predictors enables the exploration of a large chemical/structural space, facilitating the discovery of new materials. Moreover, ML enables the identification of hidden relationships in a high dimensional chemical/structural space that cannot be considered by humans. Through core-loss spectroscopy, Timoshenko et al. determined the atomic structures of nanoparticles and metals directly from the acquired spectra. The present authors also used an artificial neural network to directly extract material properties from core-loss spectra. There has been a progressive expansion of the application of core-loss spectroscopy.

In the previous study, the group of the present authors has developed an interpretation and prediction method of core-loss spectra based on hierarchical clustering and decision tree. However, the proposed method had three shortcomings that prevent an actual application to a large spectrum dataset. Firstly, the constructed decision tree tended to be very deep, which made the interpretation highly confusing. Secondly, the hierarchy of the dendrogram did not correspond to that of the decision tree, resulting in the possibility of the loss of important correlations. Finally, the method makes several clusters, resulting in ambiguating what spectral features actually correlate with the extracted descriptors.

In the present study, we have improved the method to overcome these shortcomings, and to achieve more human-like interpretations by ML. We demonstrated automatic and accurate determinations of the spectrum-structure relationships for more than 400 core-loss spectra.

The figure below shows the schematic illustration of the proposed method. The proposed method enables the automatic determination of physically and chemically reasonable spectrum–structure relationships without arbitrariness in data-driven manner, which is considerably difficult only with simulation or conventional machine leaning techniques. Such relationships are useful for understanding what structural parameters cause changes in the spectrum, providing a way for the better interpretation of spatial distributed or time evolutionary data. Furthermore, although the present work focused on the core-loss spectrum from small organic molecules, the proposed method can be readily extended to other spectral data. It is expected to contribute to a better understanding of the spectrum–structure relationship in various spectroscopy applications.

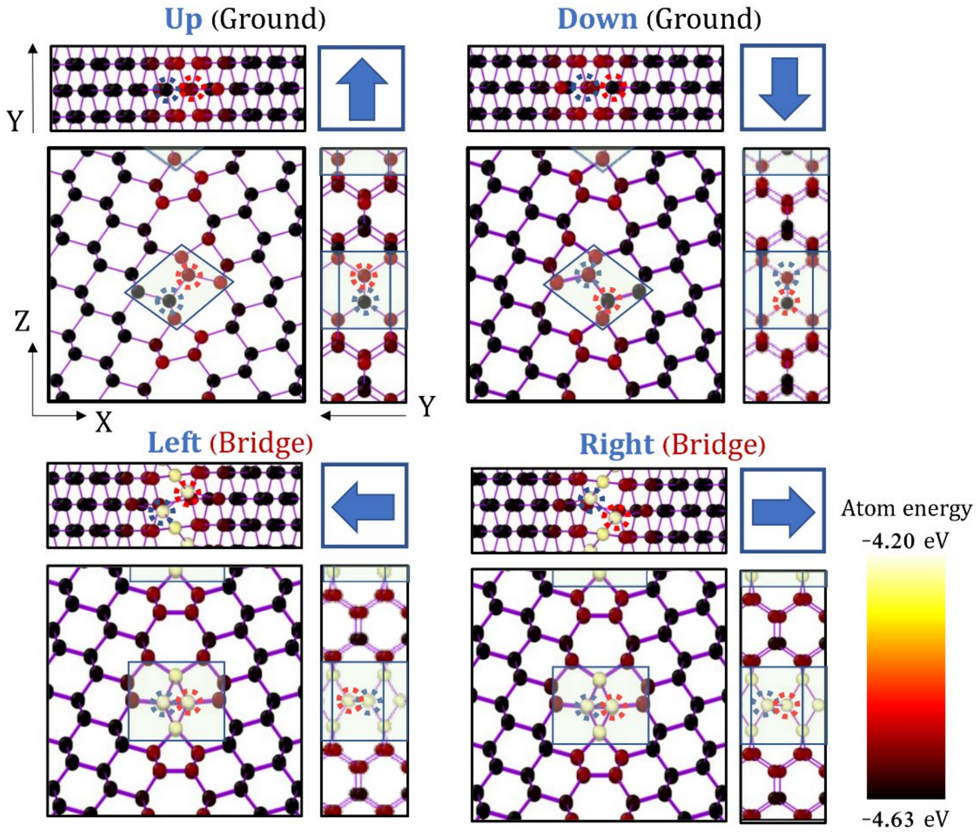


1.2 <u>A defect formation mechanism induced by structural reconstruction of a well-known silicon grain boundary</u>

Since grain boundary (GB) structures play a significant role in affecting the properties of many important polycrystalline materials, extensive studies have been done to study the structure-property relationship of GBs, where one of the main tasks is to determine their atomic structures under different physical environments. GBs in diamond-structured materials, including those in polycrystalline silicon, germanium, and diamond (carbon), have notable effects on the performances of many functional devices like solar cells and transistors, and there have been reports presenting routines to find the most stable atomic structures which are the ground states of their twist and symmetric tilt GBs at 0K by using atomic simulation. These previous studies have suggested that the ground state of a silicon GB can be obtained by structural reconstruction from its metastates which were attained by relaxing some initial configurations sampled from the configuration space of a coincident-site-lattice (CSL) GB, which can be distinguished by their microscopic parameters such as the relative rigid body translation (RBT) between the two adjacent crystals. While these explored meta-stable GBs with a reconstruction relationship to the ground state have been thought as unstable as they have higher GB energy at 0K, it is questionable to ignore their presence at elevated temperatures considering the effects of entropy triggering phase transition and of the kinetic energy bringing possibility for temporary existence of metastates. For CSL GBs in diamond-structured materials with well-defined structural units, a structural unit can perform reconstruction independently without requiring other units to do so simultaneously due to the stability of covalent bonds. Therefore, it is worth investigating the thermodynamical stability of a GB when its unit structures reconstruct non-simultaneously, making a structure consisting of domains like an Ising model. It is also important to reveal the probability of a single unit structure of the ground state to transform into a unit of a metastate, making a defect-like structure at elevated temperature. While there have been reports on segregation and vacancies at GBs, such defects by GB units reconstruction have not been reported before. Revealing this behavior can help to discover new atomic structures and properties of GBs at elevated temperatures. Also, investigating the structural reconstruction of diamond-structured GBs can provide new insights into studying the GB phase transitions and make comparisons with the previously reported behaviors of metallic GBs. Previous studies have reported that GBs consisting of metallic elemental materials have different GB phases, and GB phases of face-centered cubic (fcc) and body-centered cubic (bcc) metals were explored by atomic simulation. Furthermore, A direct observation of the coexistence of binary GB phases in copper under atomic-scale microscopy was recently achieved. Despite these delightful discoveries of the phase behaviors of elemental fcc and bcc GBs, GB phase behaviors of other species of elemental materials with non-metallic bonds are still poorly understood. It is of scientific importance to extend such studies into materials with different types of chemical bonds, such as those with covalent bonds.

In this work, we show that the well-known ground state of the a very popular GB of silicon splits into two degenerate states distinguished by an orientational feature reflected by the relative position of a pair of flag atoms inside the unit structure and by the relative rigid body translation (RBT) between the two adjacent crystals. Then we discussed the possibility of the coexistence of these two degenerate states at elevated temperatures considering both thermodynamical and kinetical aspects through molecular dynamics (MD) simulation. From the results, we propose a defect formation mechanism of this well-known GB imitating an Ising model by discussing the thermodynamical stability of the domain structures, the interaction between the Up and Down structural units (as shown in the Figure below), and the transition paths and transition rates of a single structural unit with different neighboring environments. These results provide helpful insights into understanding the structural variation of diamond-structured GBs at elevated temperatures and the differences between the phase behaviors of covalent-bonded GBs and that of metallic GBs.

Our research unrevealed a new mechanism of the structural variation of this well-known GB at elevated temperatures and made new insights into understanding the behavior of reconstructing interfaces in crystalline silicon, which can be extended to other covalent-bonded crystalline materials.



2. Research Achievements

2.1 Number of original journal papers: 10
2.2 International conference: 9 (including 5 invited presentations)
2.3 Domestic conference: 16 (including 11 invited presentation)
2.4 Number of patents: 0

3. List of awards

- 1 awards in domestic conferences, society, and university.

4. Research Grants

4.1 Total number of research grants: 3
4.2 Number of collaboration research with industries: 2

4.3 List of major research grants (serving as Principal Investigator)

- Grant-in-Aid for Scientific Research A "Materials design via atomic resolution vibrational analysis" from MEXT
- Grant-in-Aid for Scientific Research on Innovative Areas (Research in a proposed research area) "Computational design of functional core using informatics approaches" from MEXT
- CREST

5. Education

5.1 Number of Ph.D. students (including current students): 1

5.2 Number of master students (including current students): 5

5.3 Number of other students: 0

6. Publication list

Journal Papers (selected)

1. "A defect formation mechanism induced by structural reconstruction of a well-known silicon grain boundary", YS. Xie, K. Shibata, and T. Mizoguchi , Acta Mater., 250 (2023) 118827-1-11.
2. "Determination of the Spectrum–structure Relationship by Tree Structure-based Unsupervised and Supervised Learning" *S. Kiyohara, *K. Kikumasa, K. Shibata, and T. Mizoguchi, Ultramicroscopy, 233 (2022) 113438-1-8.
3. "interface_master: Python package building CSL and approximate CSL interfaces of any two lattices -- an effective tool for interface engineers", YS. Xie, K. Shibata, and T. Mizoguchi, arxiv, (2022) https://arxiv.org/abs/2211.15173
4. " Multimetastability effect on the intermediate stage of phase separation in BaO-SiO2 glass", K. Nakazawa, Y. Tsukada, S. Amma, K. Shibata, and T. Mizoguchi,, Phys. Rev. Res., 4 (2022) 033052-1-8.
5. "Ceramic Science of Crystal Defect Cores", K. Matsunaga, M. Yoshiya, N. Shibata, H. Ohta, and T. Mizoguchi,, J. Ceram. Soc. Jpn, (2022) doi.org/10.2109/jcersj2.22080.
6. "Simulated carbon K edge spectral database of organic molecules", K. Shibata, K. Kikumasa, S. Kiyohara, and T. Mizoguchi, Scientific Data, 9 (2022) 214-1-11
7. "Quantum Oscillations from Fermi arc Surface States in Cd3As2 Nanowires", Y. Miyazaki, T. Yokouchi, K. Shibata, Y. Chen, H.Arisawa, T. Mizoguchi, E. Saitoh, and Y. Shiomi, Phys. Rev. Res. 4 (2022) L022002-1-6.
8. "Determination of the Spectrum–structure Relationship by Tree Structure-based Unsupervised and Supervised Learning", *S. Kiyohara, *K. Kikumasa, K. Shibata, and T. Mizoguchi, Ultramicroscopy, 233 (2022) 113438-1-8
9. "A brute-force code searching for cell of non-identical displacement for CSL grain boundaries and interfaces", YS. Xie, K. Shibata, and T. Mizoguchi¸Comp. Phys. Comm. 273 (2022) 108260-1-8
10. "Quantification of the Properties of Organic Molecules Using Core-Loss Spectra as Neural Network Descriptors", K. Kikumasa, S. Kiyohara, K. Shibata, and T. Mizoguchi¸Advanced Intelligent Systems, 4 (2022) 2100103-1-10.

Conference Presentations (selected)

Teruyasu Mizoguchi, "Application of AI for EELS", Kamakura 2022, Kamakura, Kanagawa, 11/21, 2022

Teruyasu Mizoguchi, "Machine learning approach for interface and surface"
International Symposium on Noble and Nano Materials (ISNNM2022), Jeju, Korea, 11/14, 2022

Teruyasu Mizoguchi, "Data-driven spectral analysis for materials characterization", CIMTEC 2022, Perugia, Italy, 6/23, 2022

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DATA DESCRIPTOR

Simulated carbon K edge spectral database of organic molecules

Kiyou Shibata[1✉], Kakeru Kikumasa[1], Shin Kiyohara[1,2] & Teruyasu Mizoguchi[1✉]

Here we provide a database of simulated carbon K (C-K) edge core loss spectra of 117,340 symmetrically unique sites in 22,155 molecules with no more than eight non-hydrogen atoms (C, O, N, and F). Our database contains C-K edge spectra of each carbon site and those of molecules along with their excitation energies. Our database is useful for analyzing experimental spectrum and conducting spectrum informatics on organic materials.

Background & Summary

Carbon-based organic molecules form an immense configuration space and still have a lot of potentials for unknown functionalities in various fields. In the research and development of organic materials and their functionalities, accurate characterization of configuration and prediction of functionalities are the keys for success.

Among analytical clues for materials characterization, core-loss spectra, electron-energy loss near edge structure (ELNES) and X-ray absorption near edge structure (XANES), have been widely used as one of the most effective fingerprints for determination of local atomic structures and electronic states. These core-loss spectroscopy measure energy loss due to excitation of an electron from a core orbital and the core-loss spectra contain a partial density of states of the unoccupied states and possess useful information on atomic structure and electronic structure. Analysis of the core-loss spectrum have been performed by a comparison of measured spectra to reference spectra. Recent developments in experimental equipment and facilities have enabled core loss spectroscopy at high resolution regarding time, space, and energy. On the other hand, because of the extremely large amount of spectral data obtained, it is becoming increasingly important to establish methods for efficient and automatic analysis. Organic molecules have a variety of molecular structures, and the correlation between their structures and chemical bonds and core-loss spectral shapes is complex and has been remaining elusive.

These situations have been stimulating the development of reference spectral databases including huge variety of spectra obtained by experiments[1] and calculation[2]. Although a database of the core-loss spectra for inorganic materials has been recently developed[3], databases of the organic molecules are highly limited. Furthermore, in the field of materials science, application of informatics on predicting and designing various materials properties from spectrum data in data-driven manner has been attracting much interest. These facts represent increasing need for spectrum database of organic materials.

Here, we calculated core-loss spectra of carbon K (C-K) edge of 117,340 symmetrically unique sites in 22,155 molecules in a structure and property database of organic molecules[4,5], based on density functional theory (DFT). Our dataset provides theoretical fingerprints for analyzing experimental core-loss spectra, and also offers an opportunity for trying data-driven spectrum informatics on organic molecules.

Methods

Density functional theory calculations. The calculations of C-K edge spectra and excitation energies were carried out based on DFT by the first-principles plane-wave basis pseudopotential method using CASTEP code[6–11]. The generalized gradient approximation in a Perdew-Burke-Ernzerhof (GGA-PBE)[12] was adopted for the exchange-correlation functional. Spin polarization was not considered. For each carbon site in each molecule, an excited electronic structure was separately calculated using an on-the-fly potential of carbon. In the pseudopotential calculation, the core-hole effect can be taken into account by employing a special pseudopotential designed for the excited atom with a core-hole[11]. We consider a neutral excited state including a full core hole, *i.e.* a state with a 1 s core hole and an additional electron at an orbital corresponding to the original lowest unoccupied molecular orbital (LUMO) state, to calculate both the excitation energy and the spectral feature. The

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Automatic determination of the spectrum–structure relationship by tree structure-based unsupervised and supervised learning

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A R T I C L E I N F O

Keywords:
ELNES/XANES
Informatics
Machine learning

A B S T R A C T

Spectroscopy is widely used for the analysis of chemical, vibrational, and bonding information. Interpretations of the spectral features have been performed by comparing the objective spectra with reference spectra from experiments or simulations. However, the interpretation process by humans is not always straightforward, especially for spectra obtained from unknown or new materials. In the present study, we developed a method using machine learning techniques to obtain human-like interpretation automatically. We combined unsupervised and supervised learning methods; then applied it to the spectrum database which includes more than 400 spectra of water and organic molecules containing various ligands and chemical bonds. The proposed method has successfully found the correlations between the spectral features and descriptors of the atoms, bonds, and ligands. We demonstrated that the proposed method enabled the automatic determination of reasonable spectrum–structure relationships such as between π^* resonance in C-K edges and multiple bonds. The proposed method enables the automatic determination of physically and chemically reasonable spectrum–structure relationships without arbitrariness in data-driven manner, which is considerably difficult only with simulation or conventional machine leaning techniques. Such relationships are useful for understanding what structural parameters cause changes in the spectrum, providing a way for the better interpretation of spatial distributed or time evolutionary data. Furthermore, although the present work focused on the ELNES/XANES spectrum from small organic molecules, the proposed method can be readily extended to other spectral data. It is expected to contribute to a better understanding of the spectrum–structure relationship in various spectroscopy applications.

1. Introduction

Characterization of atomic and electronic structures is indispensable in materials chemistry. The process is necessary for the development of new materials; thus a variety of analytical techniques are employed based on the relevant system, the desired information, and the required resolution. Since chemical reactions particularly occur in very local environments, an understanding of the atomic and electronic structures is very important. Moreover, during chemical reactions, the atomic and electronic structures of molecules undergo dynamic changes depending on the reaction environment; hence an understanding of the entire transition is also imperative.

In the context of the foregoing, ELNES/XANES spectroscopy is a promising technique for the analysis of both atomic and electronic structures with nano-order spatial resolution and time resolution. The technique utilizes X-ray or fast electrons as probes; the spectral data thus reflects the atomic and electronic structures at the nano- subnano-scale in real space. It specifically offers very local information such as the constituent elements, oxidation state, coordination environment, and chemical bonds [1–6]. Furthermore, ELNES/XANES experiments can be performed in operando, in the case of which the spectrum changes also reflect the atomic and electronic structure transitions that occur during a chemical reaction. This has informed the broad use of ELNES/XANES spectroscopic techniques in the investigation of chemical materials science processes such as catalytic reaction, oxygen reduction reaction (ORR), and electrochemical reaction [7–12].

However, the interpretation of the obtained ELNES/XANES spectra is not usually straightforward. Even if distinct feature changes in the time-

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[1] S.K. and K.K. contributed equally to this work. S.K., K.K. and T.M. designed research; S.K. and K.K. performed research; S.K., K.K, K.S. and T.M. analyzed data; and S.K., K.K., K.S. and T.M. wrote the paper.

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OI http://doi.org/10.2109/jcersj2.22080

JCS Japan

REVIEW

Ceramic science of crystal defect cores

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Point defects dislocations grain boundaries and interfaces are always involved in ceramic microstructures and play important roles for physical and chemical properties of ceramics. Currently proper control of these crystal defects is inevitable to tailor ceramic materials with superior properties. This article reviews recent research projects on distinct properties and phenomena in ceramics due to crystal defects. In particular we would like to emphasize importance of central core regions of crystal defects namely crystal defect cores". They have specific electronic and atomic structures that are different from those in bulk. Recent advances of nanoscale characterizations and theoretical calculations make it possible to acquire a variety of quantitative data on electronic structures enclosed at the crystal-defect cores which gives clear understanding of various ceramic properties at the electronic and atomic levels.

Key-words : Crystal defect, DFT, Machine learning, STEM, Plastic deformation, Grain boundary migration, Thermal conductivity, Thermoelectricity, Spectroscopy

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1. Introduction

Ceramic materials are polycrystalline solids that are made up of metal and non-metal elements, and inorganic crystal grains with specific crystal structures are fundamental components in ceramic microstructures. However, atomic structures of such inorganic crystal grains in ceramic microstructures are generally not ideal, and the periodic arrays of atoms are occasionally violated by the presence of crystal defects. Crystal defects can be thermodynamically or intentionally introduced in ceramic processes of heating and cooling. Moreover, grain boundaries (GBs) or interfaces, which are planer crystal defects, are formed during fabrication processes and involved in real ceramic microstructures. Even if concentrations and volume fractions of crystal defects are in the order of a few percent, it has been recently reported that crystals defects can significantly improve or degrade overall physical and chemical properties of ceramic materials. For further advances of ceramic science and industries, therefore, it is essential to obtain detailed knowledge on nature of crystal defects in ceramics systematically and exploit them in materials developments.

Although impacts of crystal defects have long been pointed out even in the field of ceramics, conventional theories of crystal defects are still limited to crystallographic or geometrical consideration in most cases. As a matter of course, most of distinct materials properties due to crystal defects arise from specific electronic and atomic structures localized at their core regions. Electronic structures enclosed at crystal-defect cores in the ground state as well as in the excited states in response to external stimuli (mechanical, optical and electrical) should be intimately related to development of various materials properties (see **Fig. 1**). However, fundamental knowledge on such crystal-defect core regions remains insufficient. This may be mainly because it has not been easy to obtain direct information on nanoscale structures at crystal-defect cores in real ceramic materials. It is likely that preceding materials-science studies were not able to step into physics of crystal defect cores due to limited abilities of conventional experimental and theoretical techniques. In this regard, recent advances of resolution and accuracy of nanoscale characterization techniques and theoretical calculations are remarkable and make it possible to acquire not only atomic-level but also electronic-level information on crystal defects just at the cores quantitatively.

On this scientific basis, we have proposed the concept of "crystal defect cores" as a new materials science principle, and are conducting extensive research projects on ceramic materials. In this review article, our recent challenges based on this concept are described as several scientific results on electronic and atomic structures of crystal

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Multimetastability effect on the intermediate stage of phase separation in BaO-SiO_2 glass

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Controlling the phase separation phenomenon can enhance the properties of glass materials, such as transparency and strength. However, the initial and intermediate stages of phase separation of amorphous glass are yet to be understood completely. In this study, we performed an *in situ* observation on glass through scanning transmission electron microscopy, which possesses a high spatial resolution and chemical sensitivity. We visualized the phase-separated structure in the initial and intermediate stages of phase separation and observed a local and rapid change in the phase-separated structures and the formation of regions with advanced and delayed degrees of phase separation. The results were compared with the phase-field simulation and it was concluded that the characteristic change of the phase-separated structures is attributable to the multimetastability of the amorphous phase.

DOI: 10.1103/PhysRevResearch.4.033052

I. INTRODUCTION

Phase separation is a phenomenon that occurs when a uniform phase becomes unstable due to changes in temperature or pressure and separates into two stable phases [1]. This phenomenon can enhance the physical and chemical properties of glass [2] and is primarily studied in Fourier space through light [3], x-ray [4], or neutron [5] scattering methods. These studies have revealed the laws for developing average structure [3,6]. Cahn and Hilliard modeled phase separation using the Gibbs energy of the system and the composition gradient energy [7–11], and showed that phase separation proceeds to minimize the total energy. From the viewpoint of Gibbs energy, the phase separation type mainly can be classified into spinodal and binodal types, while the phase separation process can be divided into initial, intermediate, and final stages [1]. The initial stage begins with compositional fluctuation or nucleation, which becomes large in the intermediate stage and continues until the composition is binarized. After the composition is binarized, the phases begin coarsening in the final stage.

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Amorphous glass materials have random and isotropic atomic structures [12]. The atomic structures of glass have multimetastable states that can undergo structural transitions (i.e., crystallization) [13], affecting the local phase-separated structure. The effect of the phase-separated structure on crystallization [14] and the cessation of the coarsening of the phase-separated structure have been reported [15]. Moreover, the effect on the local phase-separated structure when the transition occurs in the final stage has been relatively well studied [14–17]. However, the effect on the initial and intermediate stages has not been extensively researched and only a few observations of the phase-separated structure in the initial and intermediate stages have been made by imaging [18]. Because the phase-separated structure formation proceeds mainly during the initial and intermediate stages of the phase separation, it is essential to gain insight into these stages from the perspective of the phase separation process by imaging. Moreover, imaging the formation of the phase-separated structure is important to obtain guidelines for controlling the phase-separated structure and crystallization. The nanoscale sizes of the phases and the slight compositional differences between the phases in the initial and intermediate stages made it challenging to make observations through imaging. High resolution and chemical sensitivity are required for imaging the phase-separated structure at the initial and intermediate stages. Recently, our group revealed that the high-angle annular dark-field (HAADF) method conducted by scanning transmission electron microscopy (STEM) possesses sufficient spatial resolution and chemical sensitivity to observe the

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Full length article

A defect formation mechanism induced by structural reconstruction of a well-known silicon grain boundary.

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A R T I C L E I N F O

Keywords:
Grain boundaries
Interface phase transition
Molecular dynamics
Ising model
Diamond-structured materials

A B S T R A C T

Diamond-structured grain boundaries (GBs) are important because they have notable effects on the performances of many functional devices. Previous studies have suggested that the most stable structures of some silicon GBs can be obtained by structural reconstruction from some meta-stable GBs explored at 0 K by atomic simulation. While GB reconstruction is possible to enable these meta-stable GBs to exist at elevated temperatures, reports on such behaviors are rare. This work unveiled a non-reported GB reconstruction from two degenerate ground-states of a well-known silicon GB which can be distinguished by an orientational feature of their unit structures. The reconstructing structures were verified stable by density-functional-theory (DFT) simulation. By thermodynamical and kinetical discussion, we have shown that the structural variation of this well-known GB at elevated temperatures is more likely to be dominated by this reconstruction mechanism rather than by transforming to other metastates. Such a reconstruction mechanism allows the whole GB system to be treated as an Ising model with a second-ordered phase transition. By applying harmonic transition state theory, we predicted the possible concentration of the defects induced by reconstruction at elevated temperatures and discussed their effects on the band structure of the GB by DFT simulation. An explanation was made on the cause of the difference between the phase transition behavior of this silicon GB and that of a reported copper GB. Our research made new insights into understanding the behavior of reconstructing interfaces in covalent-bonded crystalline materials.

1. Introduction

Since grain boundary (GB) structures play a significant role in affecting the properties of many important polycrystalline materials [1,2], extensive studies have been done to study the structure-property relationship of GBs, where one of the main tasks is to determine their atomic structures under different physical environments. GBs in diamond-structured materials, including those in polycrystalline silicon, germanium, and diamond (carbon), have notable effects on the performances of many functional devices like solar cells and transistors [3–5], and there have been reports presenting routines to find the most stable atomic structures which are the ground states of their twist and symmetric tilt GBs at 0 K by using atomic simulation [6–8]. These previous studies have suggested that the ground state of a silicon GB can be obtained by structural reconstruction from its metastates which were attained by relaxing some initial configurations sampled from the configuration space of a coincident-site-lattice (CSL) GB, which can be distinguished by their microscopic parameters such as the relative rigid body translation (RBT) between the two adjacent crystals [9]. While these explored meta-stable GBs with a reconstruction relationship to the ground state have been thought as unstable as they have higher GB energy at 0 K, it is questionable to ignore their presence at elevated temperatures considering the effects of entropy triggering phase transition and of kinetic energy bringing possibility for temporary existence of metastates. For CSL GBs in diamond-structured materials with well-defined structural units, a structural unit can perform reconstruction independently without requiring other units to do so simultaneously due to the stability of covalent bonds. Therefore, it is worth investigating the thermodynamical stability of a GB when its unit structures reconstruct non-simultaneously, making a structure consisting of domains like an Ising model. It is also important to reveal the probability of a single unit structure of the ground state to transform into a unit of a metastate, making a defect-like structure at elevated temperature. While there have been reports on segregation and vacancies at GBs [10,11], such defects by GB reconstruction have not been reported before. Revealing this behavior can help to discover new atomic structures and properties of GBs at elevated temperatures. Also, investigating the structural reconstruction of diamond-structured GBs can provide

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NOMURA Laboratory

Current Research Activities 2022-2023

Nomura Laboratory

1. Research Topics

1.1 Hydrodynamic thermal phonon transport in graphite

Phonon hydrodynamics has been intensively reviewed owing to its peculiar phenomena, very similar to fluid dynamics. In graphite, the dominance of momentum-preserved normal scattering promoted the demonstration of second sound and phonon Poiseuille flow latterly. However, the impact of isotope scattering on the phonon hydrodynamic conduction in graphite remains vacant.

We experimentally demonstrate that phonon Poiseuille flow only exists in ^{13}C isotopically purified graphite ribbons. We investigate phonon Poiseuille flow (illustrated in Fig.1a) in suspended graphite ribbons (Fig. 1b) with both natural (1.1%) and enriched (0.02%) carbon isotope concentrations. Based on a steady-state μTDTR setup, we first study the suppression of thermal conductivity (κ) by isotope-phonon scattering, as shown with a significant reduction of κ in a natural graphite ribbon at the intermediate temperature range in Fig. 1c. Furthermore, we examine the observation of phonon Poiseuille flow under an explicit criterion ($\kappa/G_{ballistic}$) in Fig. 1d. As the temperature increases, κ is enhanced over the ballistic case from 30 to 60 K, attributed to the hydrodynamic transport of phonons in the isotopically enriched sample. Whereas $\kappa/G_{ballistic}$ solely decreases in the natural graphite sample, which indicates the absence of phonon Poiseuille flow resulted from the sufficient momentum-destroyed isotope scattering [1]. Supported by our theoretical calculation by solving the phonon Boltzmann transport equation, we demonstrate that the phonon Poiseuille flow can only be observed in isotopically purified graphite ribbon, thus orientating future progress for a deeper understanding of phonon hydrodynamics in solids [2].

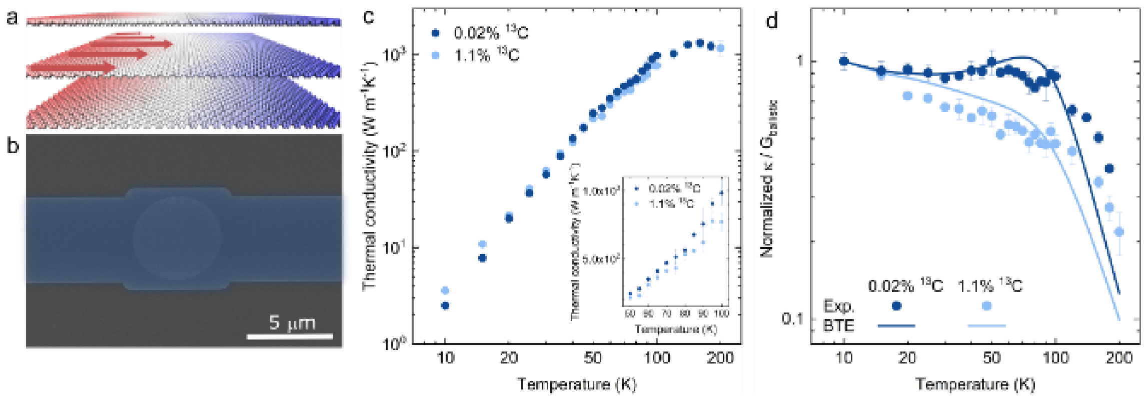

Figure 1: (a) Illustration of phonon Poiseuille flow in graphite. (b) SEM image of a 65-nm-thick suspended graphite ribbon (blue color). (c) Temperature-dependent thermal conductivity of isotopically enriched (dark blue) and natural (light blue) graphite ribbons. I (d) Normalized thermal conductivity over ballistic thermal conductance as a function of temperature.

References: [1] X. Huang, te al. Nat. Commun. 14, 2044 (2023). [2] Y. Guo, et al., Phys. Rev. B 104, 075450 (2021).

1.2 Thermal transport by surface phonon polaritons in SiN nanofilms

Thermal conduction becomes less efficient as structures scale down into submicron sizes due to the predominant phonon-boundary scattering that hinders phonons more efficiently than Umklapp scattering. Recent studies indicated that this thermal performance reduction could be avoided by using surface phonon-polaritons (SPhPs), which are evanescent electromagnetic waves generated by the hybridization of optical phonons and photons. These waves propagate along the surface of polar dielectric materials and could be heat carriers capable of remarkably enhancing the thermal performance of micro- and nanoscale devices. We experimentally observe the dominant heat contribution of SPhPs in SiN nanofilms

Figure 1 shows the measurement setup and thermal conductivity of four SiN nanomembranes (thickness: 30-200 nm) at different temperatures ranging from 300 and 800 K. The thermal conductivity of the membranes thinner than 50 nm increases with temperature, as expected for the SPhP contribution, while that of a 200 nm-thick membrane decreases, in agreement with the phonon counterpart (Fig. 1c). In general, the thermal conductivity decreases as the temperature increases due to more frequent phonon-phonon scattering. However, the two thinner membranes, i.e. 30 and 50 nm-thick membranes, become twice more conductive when their temperature rises from 300 to 800 K [1]. This behavior represents the fingerprints of SPhPs: much longer propagation length of SPhPs ~ mm, while the mean free paths of phonons are below 100 nm [2]. The hybridization of phonons with photons enables ultrafast thermal transport in such a system.

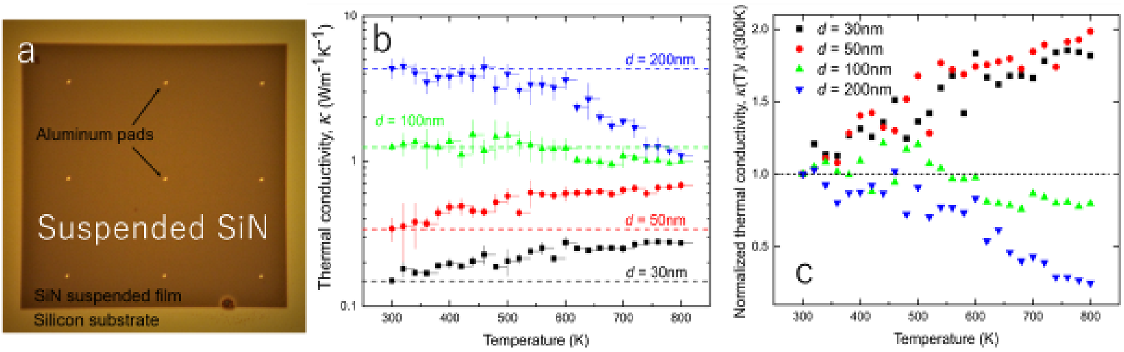

Fig. 1. (a) Optical microscope image of the sample. (b) Thermal conductivity as a function of temperature, for SiN membranes with different thicknesses. (c) Thermal conductivity normalized by the value at room temperature.

References

[1] Y. Wu et al., Sci. Adv. 6, eabb4461 (2020). [2] Y. Wu, et al., Appl. Phys. Lett. 121, 112203 (2022).

2. Research Achievements

2.1 Number of original journal papers: 16

2.2 International conference: 32 (including 13 invited presentations),

2.3 Domestic conference: 29 (including 7 invited presentation)

2.4 Number of patents: 1

3. List of awards

1. R. Yanagisawa, Best Paper Award, PowerMEMS 2022 (IEEE)
2. S. Koike, 6th Phonon Engineering Workshop, Excellent Poster Award

4. Research Grants

4.1 Total number of research grants: 14

4.2 Number of collaboration research with industries: 6

4.3 List of major research grants (serving as Principal Investigator)

- CREST, JST
- Research Grants by Japan Society for the Promotion of Science (KAKENHI)

5. Education

5.1 Number of Ph.D. students (including current students): 5

5.2 Number of master students (including current students): 2

5.3 Number of other students: 0

6. Publication list (Selected)

Journal Paper

1. S. Volz, M. Nomura, and J. Ordonez-Miranda, "Resonant polariton thermal transport along a vacuum gap," Phys. Rev. Appl. 18, L051003 (2022).
2. S. Tachikawa, J. Ordonez-Miranda, Y. Wu, L. Jalabert, R. Anufriev, S. Volz, and M. Nomura, "In-plane surface phonon-polariton thermal conduction in dielectric multilayer systems," Appl. Phys. Lett., 121, 202202 (2022).
3. X. Huang, Y. Guo, S. Volz, and M. Nomura, "Mapping phonon hydrodynamic strength in micrometer-scale graphite structures," Appl. Phys. Express, 15 105001. (2022)
4. Y. Wu, J. Ordonez-Miranda, L. Jalabert, S. Tachikawa, R. Anufriev, H. Fujita, S. Volz, and M. Nomura, "Observation of heat transport mediated by the propagation distance of surface phonon-polaritons over hundreds of micrometers," Appl. Phys. Lett. 121, 112203 (2022).
5. J. Ordonez-Miranda, R. Anufriev, M. Nomura, and S. Volz, "Net heat current at zero mean temperature gradient," Phys. Rev. B 106, L100102 (2022).
6. J. Ordonez-Miranda, Y. Wu, M. Nomura, and S. Volz, "Near-isotropic polariton heat transport along a polar anisotropic nanofilm," iScience 25, 104857 (2022).

7. M. Nomura, V. Laude, and M. Maldovan, “Phononic Crystals at Various Frequencies,” APL Mater. 10, 050401 (2022).
8. T. Sato, Z. Milne, M. Nomura, N. Sasaki, R. Carpick, and H. Fujita, “Ultrahigh Strength and Shear-Assisted Separation of Sliding Nanocontacts Studied in situ,” Nat. Commun. 13, 2551 (2022).
9. Z. Zhang, Y. Guo, M. Bescond, J. Chen, M. Nomura, and S. Volz, “How coherence is governing diffuson heat transfer in amorphous solids,” npj Comput Mater. 8, 96 (2022).
10. R. Anufriev, Y. Wu, J. Ordonez-Miranda, and M. Nomura, “Nanoscale limit of the thermal conductivity in crystalline silicon carbide membranes, nanowires, and phononic crystals,” NPG Asia Mater. 14, 35 (2022).
11. M. Nomura, R. Anufriev, Z. Zhang, J. Maire, Y. Guo, R. Yanagisawa, and S. Volz, “Review of thermal transport in phononic crystals,” Mater. Today Phys. 22, 100613 (2022). (Invited review)
12. Z. Zhang, Y. Guo, M. Bescond, J. Chen, M. Nomura, and S. Volz, “Heat conduction theory including phonon coherence,” Phys. Rev. Lett. 128, 015901 (2022).
13. R. Anufriev and M. Nomura, “Phonon Engineering for Quantum Hybrid Systems,” Quantum Hybrid Electronics and Materials, 15-24, Springer Nature (2022).

International Conference

1. M. Nomura (Invited), “Power density enhancement by thermal design of a planar-type Si TEG,” Thermal Control, Unusual behaviors in electron and lattice thermal conductivity online workshop, Invited Talk 1, online.
2. M. Nomura (Invited), “Planar-type double-cavity Si thermoelectric generators,” WPI-MANA International Symposium 2022, Day2-7-2, Tsukuba, Japan (2022).
3. R. Anufriev(invited), Y. Wu, S. Gluchko, S. Volz, and M. Nomura, “Ballistic phonon and thermal transport at nanoscale,” Plasmons and Vibrational Dynamics in Nanomaterials Seminar, Poznan, Poland (2022).
4. M. Nomura (Invited), “Nanophonoic Si thermoelectric devices with phonon engineering,” IEEE International Nanodevices & Computing (INC) Conference 2022, online (2022).
5. M. Nomura (Invited), and X. Huang, “Hydrodynamic phonon transport in graphite micro ribbons,” International Conference on Thermodynamics and Thermal Metamaterials 2022, A2-1, online (2022).
6. R. Anufriev (Invited), A. Ramiere, J. Maire, S. Gluchko, J. Ordonez-Miranda, S. Volz, and M. Nomura, “Ballistic heat conduction at nanoscale: demonstrations and applications,” Colloquium at Los Alamos National Laboratory, USA, online (2022).

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Review of thermal transport in phononic crystals

Masahiro Nomura [a, b, *], Roman Anufriev [a], Zhongwei Zhang [a], Jeremie Maire [c], Yangyu Guo [a], Ryoto Yanagisawa [a], Sebastian Volz [a, d]

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A B S T R A C T

Thermal transport at the nanoscale level is attracting attention not only because of its physically interesting features such as the peculiar behavior of phonons due to their pronounced ballistic and wave-like properties but also because of its potential applications in alleviating heat dissipation problems in electronic and optical devices and thermoelectric energy harvesting. In the last quarter-century, researchers have elucidated the thermal transport properties of various nanostructured materials, including phononic crystals (PnCs): artificial periodic structures for phonons. PnCs are excellent platforms for investigating thermal transport owing to their well-defined structural parameters. In addition, it is interesting to control thermal transport by interference, as demonstrated in the low-frequency regime with elastic waves and sounds. In this article, we focus on high-frequency phonons and review the thermal transport in semiconductor PnCs. This comprehensive review provides an understanding of recent studies and trends, organized as theoretical and experimental, in terms of the quasiparticle and wave aspects.

1. Introduction

A phonon is a collective excitation in condensed matter, which is a quantization of the vibration mode in elastic structures. Although phonons are not as popular as photons or electrons, they are ubiquitous in our daily lives. Phonons have a broad spectrum and are labeled in different frequency ranges as sound waves ($10-10^4$ Hz), ultrasound (10^4-10^8 Hz), hypersound (10^8-10^{11} Hz), and heat (over 10^{11} Hz). Phonons in the lower frequency range are often excited electrically in the GHz range and are used in various applications, including musical instruments, measurements, sensing, imaging, and piezoelectric transducers. Micro- or nano-electromechanical systems, known as MEMS/NEMS, are exemplary applications of well-defined phonons generated in artificial mechanical structures [1,2]. In the very-high-frequency range, although the boundary is unclear, the thermal vibrations of atoms or molecules are the sources of phonons, called thermal phonons.

It is challenging and interesting to pursue the possibility of reducing or increasing the thermal conductivity by nanostructuring. The phonon propagation can be engineered by using phononic crystals (PnCs)—artificial materials with a periodic structure—for phonon interference owing to the induced periodicity. PnCs are often used to investigate or control sound waves, acoustic waves, and even heat transfer in solids. There are many insightful books and reviews on phonon transport in PnCs in various frequency ranges [3–8]; we focus on PnCs in the thermal spectral range in this review.

Fig. 1 shows an increasing number of articles on PnCs and thermal transport in PnCs, published since 2005. The percentage of articles on thermal transport in terms of the articles on PnCs also shows an increasing trend. The dimensions of PnCs used for heat conduction engineering are typically at the scale of the thermal phonon mean free path (MFP), the average distance between the scatterings, or the thermal phonon wavelength depending on the phenomenon of interest. Fig. 2 shows the different types of PnC structures for thermal transport studies in the incoherent and coherent regimes.

In the incoherent thermal transport regime, materials with high thermal conductivity and established process technologies, such as

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ARTICLE

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Ultrahigh strength and shear-assisted separation of sliding nanocontacts studied in situ

Takaaki Sato[1✉], Zachary B. Milne[2], Masahiro Nomura[3], Naruo Sasaki[4], Robert W. Carpick[1] & Hiroyuki Fujita[3,5]

The behavior of materials in sliding contact is challenging to determine since the interface is normally hidden from view. Using a custom microfabricated device, we conduct in situ, ultrahigh vacuum transmission electron microscope measurements of crystalline silver nanocontacts under combined tension and shear, permitting simultaneous observation of contact forces and contact width. While silver classically exhibits substantial sliding-induced plastic junction growth, the nanocontacts exhibit only limited plastic deformation despite high applied stresses. This difference arises from the nanocontacts' high strength, as we find the von Mises stresses at yield points approach the ideal strength of silver. We attribute this to the nanocontacts' nearly defect-free nature and small size. The contacts also separate unstably, with pull-off forces well below classical predictions for rupture under pure tension. This strongly indicates that shearing reduces nanoscale pull-off forces, predicted theoretically at the continuum level, but not directly observed before.

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ARTICLE Open Access

Nanoscale limit of the thermal conductivity in crystalline silicon carbide membranes, nanowires, and phononic crystals

Roman Anufriev[1], Yunhui Wu[1], Jose Ordonez-Miranda[2] and Masahiro Nomura[1]

Abstract
Silicon carbide (SiC) aims to be the number one material for power microelectronics due to its remarkable thermal properties. Recent progress in SiC technology finally enabled the fabrication of crystalline SiC nanostructures. Yet, the thermal properties of SiC at the nanoscale remain overlooked. Here, we systematically study heat conduction in SiC nanostructures, including nanomembranes, nanowires, and phononic crystals. Our measurements show that the thermal conductivity of nanostructures is several times lower than that in bulk and that the values scale proportionally to the narrowest dimension of the structures. In the smallest nanostructures, the thermal conductivity reached 10% of that in bulk. To better understand nanoscale thermal transport in SiC, we also probed phonon mean free path and coherent heat conduction in the nanostructures. Our theoretical model links the observed suppression of heat conduction with the surface phonon scattering, which limits the phonon mean free path and thus reduces the thermal conductivity. This work uncovers thermal characteristics of SiC nanostructures and explains their origin, thus enabling realistic thermal engineering in SiC microelectronics.

Introduction

The interplanetary ambitions of our civilization demand electronics working under extreme conditions of outer space and distant planets. High temperatures, intense radiation, and toxic environments are only a few of the challenges waiting for our spacecrafts on Venus or Jupiter. These challenges call for electronics powered by exceptionally resistive materials. Silicon carbide (SiC) is the material that fits the mission like no other. Tolerant to high temperatures, resistant to radiation, and chemically inert, SiC has been named "Tougher than Hell"[1] for its excellent material properties. The flip side of SiC has always been difficult fabrication technology. For decades, researchers have struggled to obtain high-quality crystalline SiC[2–4] and to adapt the top-down fabrication technology to produce micro- and nanostructures[3]. Today, the industry has almost completely mastered the fabrication of SiC nanostructures[5,6]

Yet, despite the progress in SiC nanofabrication, the nanoscale thermal properties of SiC remain overlooked. Albeit an excellent thermal conductor at the macroscale, SiC has yet to prove its ability to dissipate heat in nanostructures. For comparison, extensive research on Si shows that the thermal conductivity of nanostructures is several times lower than that of the bulk Si[7,8]. This reduction in thermal conductivity lies at the core of the overheating problem in modern Si-based microelectronics[9,10]. Thus, knowledge of nanoscale thermal properties is essential for engineering future power electronics based on SiC. However, no systematic thermal measurements have been carried out on SiC nanostructures.

Here, we aim to demonstrate how the thermal conductivity of crystalline SiC scales with the size of the structure. We systematically measure the thermal conductivity of various SiC nanostructures and correlate it to the surface scattering of phonons. Moreover, we

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ARTICLE OPEN

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How coherence is governing diffuson heat transfer in amorphous solids

Zhongwei Zhang[1]✉, Yangyu Guo[1], Marc Bescond[2,3], Jie Chen[4]✉, Masahiro Nomura[1] and Sebastian Volz[1,2,4]✉

Thermal transport in amorphous materials has remained one of the fundamental questions in solid state physics while involving a very large field of applications. Using a heat conduction theory incorporating coherence, we demonstrate that the strong phase correlation between local and non-propagating modes, commonly named diffusons in the terminology of amorphous systems, triggers the conduction of heat. By treating the thermal vibrations as collective excitations, the significant contribution of diffusons, predominantly relying on coherence, further reveals interesting temperature and length dependences of thermal conductivity. The propagation length of diffuson clusters is found to reach the micron, overpassing the one of propagons. The explored wavelike behavior of diffusons uncovers the unsolved physical picture of mode correlation in prevailing models and further provides an interpretation of their ability to transport heat. This work introduces a framework for understanding thermal vibrations and transport in amorphous materials, as well as an unexpected insight into the wave nature of thermal vibrations.

npj Computational Materials (2022)8:96 ; https://doi.org/10.1038/s41524-022-00776-w

INTRODUCTION

Heat conduction in different states of matter exhibits diverse characteristics along with different modalities of lattice vibration, whereas driving performances in several devices and materials related fields impacted by thermal management or energy conversion. While an adequate understanding of phonons in crystals is well established, a debate still exists regarding the physical pictures of thermal vibrations and transport in amorphous materials[1–6]. Owing to the loss of the long-range lattice periodicity, the concept of phonons becomes invalid in amorphous materials and the application of the phonon-gas model for thermal transport is accordingly failing[7–11]. The limited comprehension of thermal vibrations in amorphous materials has hindered their advanced applications. In the past decades, diverse attempts have proposed theoretical frames to describe thermal vibrations and transport in amorphous materials[7–9,12–15]. Particularly, Allen and Feldman[7,8,16] established a classification of lattice vibrations into propagons, diffusons and locons. Propagons correspond to low-frequency propagating waves and locons to high-frequency localized states, while diffusons designate local modes characterized by intermediate frequencies. Their further studies demonstrated that the off-diagonal terms of the group velocity operator plays a critical role in the transport of diffusons[8,9,17]. In a recent work, Isaeva et al.[13] developed a quasi-harmonic Green-Kubo (QHGK) model to study the thermal transport in amorphous materials by including a scattering correlation between different modes. The Allen-Feldman and QHGK models share the common conceptual basis of mode correlation.

On the other hand, the study of the physical picture of thermal vibrations in amorphous materials has also attracted significant attention. In both Allen-Feldman and QHGK models, thermal vibrations are treated as plane waves in which the normal modes and scatterings are obtained from the harmonic and anharmonic lattice dynamic approaches, respectively[9,13,16]. The utilized normal modes and ill-defined group velocity[10] raise questions on the fundamentals of Allen-Feldman and QHGK descriptions.

In addition, in analogy to liquids, the collective excitations are experimentally observed and proposed to understand thermal vibrations, in analogy to liquids[18–23]. Moon et al.[20,21] found that the thermal transport in amorphous silicon (a-Si) is dominated by acoustic collective excitations rather than by normal modes and that the elastic scattering predominates. The obtained variation of lifetimes is well consistent with the frequency of the boson peak and the Ioffe-Regel crossover[18,21]. A theoretical model that can assess the thermal transport in amorphous materials by directly incorporating the collective excitations, however, is still missing. In addition, the controversies regarding the dependences of thermal conductivity (κ) on temperature and length remain topics of debate[23–27].

In this work, we theoretically investigate thermal transport in amorphous silicon, in which thermal vibrations are simultaneously interpreted as the composition of particlelike and wavelike components. The strong phase correlation or in other terms, the coherence between several diffusons, is firstly studied from wave-packet simulations and the behaviors of propagons and diffusons are clearly discriminated. Then, we apply our recently developed coherence heat conduction model[28] by estimating mode lifetimes and coherence times. The significant contribution of phase correlation to thermal conductivity is uncovered. The estimated propagation length of coherent diffusons is found beyond the one of propagons. We finally demonstrate a fundamental, unforeseen and accurate point of view to understand thermal vibrations and transport in amorphous materials.

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Heat Conduction Theory Including Phonon Coherence

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[2]*Laboratory for Integrated Micro and Mechatronic Systems, CNRS-IIS UMI 2820, The University of Tokyo, Tokyo 153-8505, Japan*
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Understanding and quantifying the fundamental physical property of coherence of thermal excitations is a long-standing and general problem in physics. The conventional theory, i.e., the phonon gas model, fails to describe coherence and its impact on thermal transport. In this Letter, we propose a general heat conduction formalism supported by theoretical arguments and direct atomic simulations, which takes into account both the conventional phonon gas model and the wave nature of thermal phonons. By naturally introducing wave packets in the heat flux from fundamental concepts, we derive an original thermal conductivity expression including coherence times and lifetimes. Our theory and simulations reveal two distinct types of coherence, i.e., intrinsic and mutual, appearing in two different temperature ranges. This contribution establishes a fundamental frame for understanding and quantifying the coherence of thermal phonons, which should have a general impact on the estimation of the thermal properties of solids.

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Phonons, i.e., quanta of vibrational waves, are commonly considered as one of the fundamental quasiparticles, simultaneously exhibiting wave- and particlelike characteristics in nanostructured crystals or bulk materials. The wavelike behavior of phonons impacts thermal properties via coherence mechanisms, as highlighted by several pioneering [1,2] and recent works [3,4]. The particlelike behavior has been treated by the Boltzmann transport equation (BTE) and the phonon-gas model in most solids [5–10]. Experiments [1,2,11–13] have revealed, however, that the wave nature of thermal phonons plays a substantial role in thermal transport, as, for example, in the observations of coherent thermal transport in nanophononic crystals [1,2,11,12]. Later, theoretical and simulation studies [14–18] were devoted to the understanding of phonon coherence, such as the one producing band folding [19–21], but missing the particle behavior. Recently, the theoretical study [22] revealed that the realistic phonon dynamics can only be manifested if both intrinsic coherence relevant to the extension of phonon wave packets and the particlelike behavior of thermal phonons are taken into account.

The conventional BTE also fails in complex crystals, as a pure particle picture cannot yield a complete description of thermal conductivity, such as in Tl_3VSe_4 [23,24]. Recently, Simoncelli *et al.* [3] developed a theory for thermal transport in glasses and complex crystals, in which the coherence between densely packed phonon branches contributes to thermal transport. A similar approach has been developed by Isaeva *et al.* [4] as well, at the same time. This mutual coherence among branches is identified as an additional phonon wave-relevant term [25–27]. The picture of this mutual coherence, which might be compared to a hopping process, however, remains physically unclear. Finally, quantifying the full coherence of thermal phonons and its effect on heat conduction remains a critical issue in transport physics.

In this Letter, a general heat conduction theory is proposed to establish an original expression for the thermal conductivity that includes the full coherent nature of phonon excitations. This expression involves both phonon lifetimes and coherence times. Those are obtained by tracking the real phonon dynamics and using a wavelet transform of the atomic trajectories during an equilibrium molecular dynamic (EMD) simulation. We show that the predictions of our theory yield significant differences from those of the conventional phonon-gas model, as demonstrated in the Tl_3VSe_4 case [see atomic structure in Fig. 1(a)]. We find that there are two types of coherence, i.e., intrinsic and mutual, which take a critical role over different temperature regions. These conclusions open unexpected insights on the reality of thermally activated phonon modes and the importance of the diverse coherence mechanisms when assessing thermal properties.

The thermal conductivity (κ) can be calculated based on the Green-Kubo approach with the autocorrelation of the heat flux $\mathbf{S}(t)$ as [28]

$$\kappa = \frac{V}{3k_B T^2} \int \langle \mathbf{S}(t) \cdot \mathbf{S}(0) \rangle dt, \tag{1}$$

where V corresponds to the system volume, k_B is the Boltzmann constant, and T is the temperature. We now

TIXIER-MITA Laboratory

Current Research Activities 2022-2023

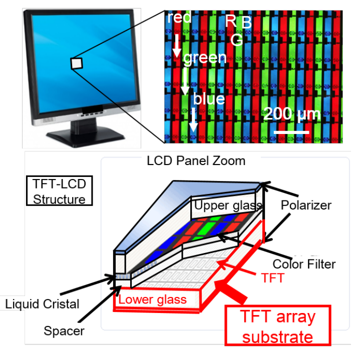
Tixier-Mita Laboratory

1. Research Topics

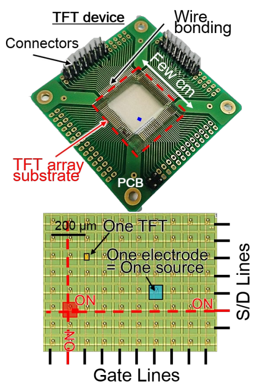
1.1 Electrophysiology of Pancreatic β cell using Thin-Film-Transistor based bio-sensing platform

The purpose of this research is to develop a bio-sensing platform based on TFT technology to study and elucidate the pathophysiology of pancreatic islet of Langerhans related diseases. Islets of Langerhans are a functional unit of the pancreas and plays a vital role in regulating glycaemia. It is composed for most part of β cell which secrete insulin at the right moment and at an appropriate amount according to glucose concentration in blood. Dysfunction of the β cells perturbs glucose homeostasis leading to life-threatening diabetes. One technique to study these islets is by measuring the action potential released by the β cell in function of glucose concentration.

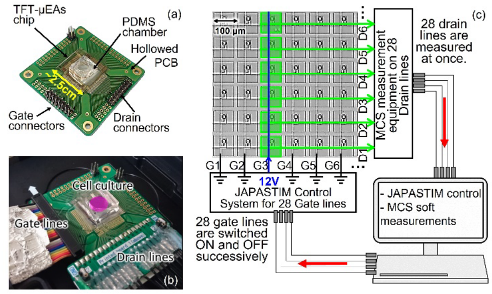
Here the activity of a β cell culture is monitored according to glucose concentration using a Thin-Film-Transistor (TFT) platform. With such platform, a high spatial resolution cartography of the activity can be obtained. In addition, the device is transparent allowing simultaneous optical observation and electrical measurement of the activity. Similar experiments were performer using commercially available Multi-Electrode-Array (MEAs), which provide also sensor array, but with much lower spatial resolution.



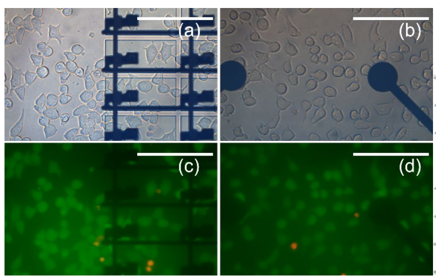
TFT array substrate description



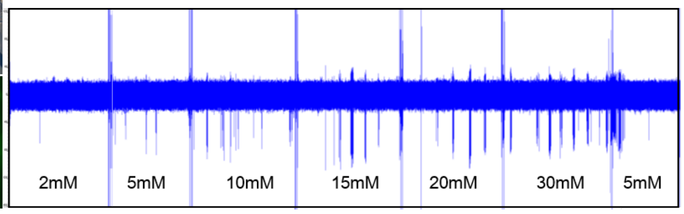
Mounted substrate and microscopic view



Set-up of for electrophysiology measurements



Optical observation of pancreatic β cell culture in bright field (a, b) and fluorescence (c, d). (a, c): the cells are cultured on TFT platform. (b, d), the cells are cultured on MEAs.

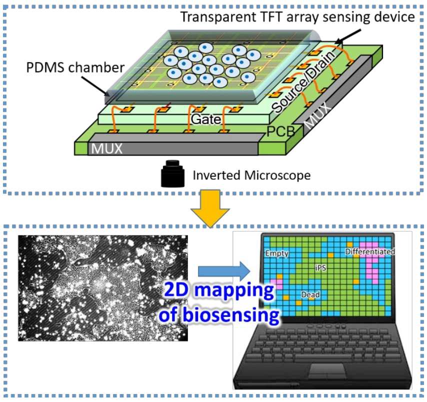


Electrophysiology measurement of the activity of pancreatic β cell cultured on a TFT platform, for different glucose concentrations.

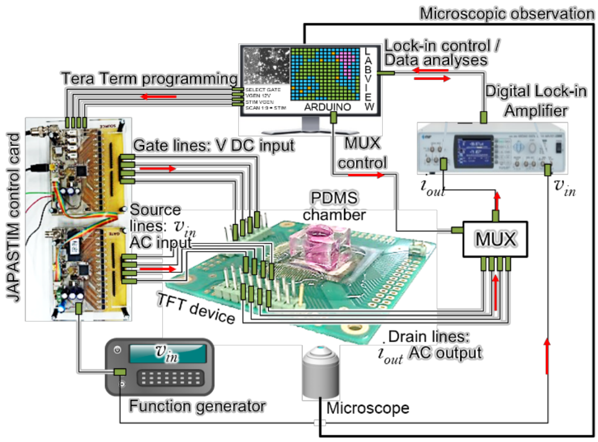
This work was performed in collaboration with Prof. Sakai from the Chemical System Engineering Dpt., The University of Tokyo, and Prof. Komori from Kindai University.

1.2 2D Bioimpedance monitoring with a TFT bio-sensing platform

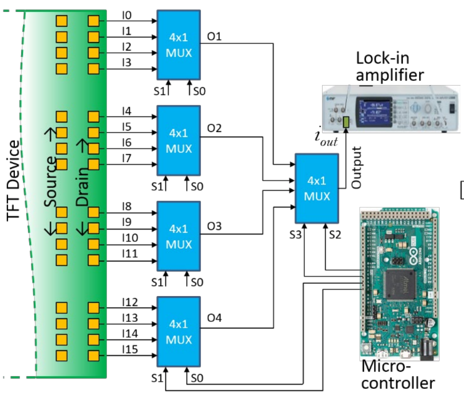
Bioimpedance sensing is a dye-free technique used to characterize electrically cells. For instance, by measuring the spatial variation of impedance, it is possible to distinguish areas with no cells, dead cells, healthy cells and so on. In this project, a Thin-Film-Transistor platform is used to monitor cell culture on long term by realizing in real time 2D cartography of bioimpedance. The platform consists in a high spatial resolution sensor matrix. The sensor matrix is connected to a matrix of switch transistors for control and selection of individual sensors. The control of the switch transistors is performed by a control card previously developed, called JAPASTIM. At the same time that card is also used to apply an input voltage to the sensor required for impedance measurement. However, JAPASTIM can't be used for the measurement of the output current signal. So, this time, a multiplexer has been fabricated to manage the output current signal. Impedance magnitude and phase are obtained based on the input voltage and output current signals measurement, on a 3x2 matrix, for air, Deionized Water and phosphate buffer conditions.



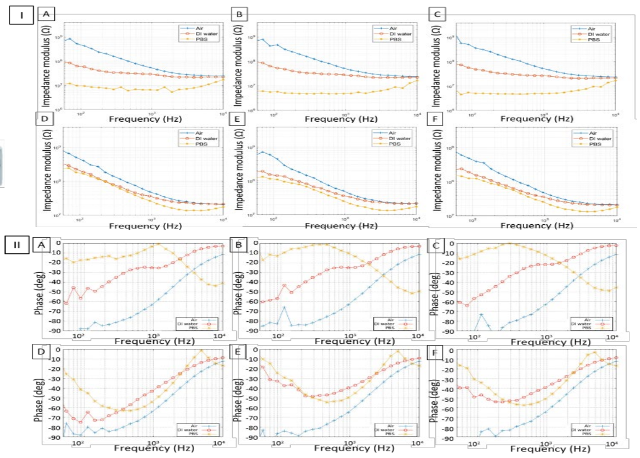
Target of the research topics. With the TFT platform, optical and electrical measurements can be obtained simultaneously.



Experimental set-up for 2D bioimpedance mapping. JAPASTIM card on the left side controls the switch transistors and applies external exciation signal to the source lines. The drain lines are connected to a multiplexer, which sends an output signal to the lock-in amplifier LI5655. The electrical monitoring can be confirmed by microscopic observation.



Scheme of the realized multiplexer circuit and control.



Impedance modulus spectrum (top: I.) and phase spectrum (bottom: II.) for air (blue), DI water (red) and PBS solution (yellow), for 3x2 sensors A, B, C, D, E and F. (σ_{AIR}= 50 aS/cm , σ_{DI}= 61 nS/cm, σ_{PBS}= 18 mS/cm).

2. Research Achievements

2.1 Number of original journal papers: 2

2.2 International conference: 1

2.3 Domestic conference: 5 (including 3 invited presentations)

2.4 Number of patents: 0

3. List of awards

- /

4. Research Grants

4.1 Total number of research grants: 1

4.2 Number of collaboration research with industries: 1

4.3 List of major research grants (serving as Principal Investigator)

1. 科研費挑戦的研究（萌芽）：代表「TFT 電極アレイを応用した高時空間分解マルチバイオイメージング法の検証」、2021. 10－2023. 03

5. Education

5.1 Number of Ph.D. students (including current students): 1

5.2 Number of master students (including current students): 0

5.3 Number of other students: 1 (1 internship student)

6. Publication list

Journal Papers (selected)

[1] Dongchen Zhu, Anne-Claire Eiler, Satoshi Ihida, Yasuyuki Sakai, Hiroshi Toshiyoshi, Agnès Tixier-Mita, Kikuo Komori, "Real-time High-resolution Measurement of Pancreatic β Cell Electrophysiology Based on Transparent Thin-film Transistor Microelectrode Arrays", IEEJ Transactions on Sensors and Micromachines, Vol. 142(10), pp. 266-272, (October 2022). DOI: https://doi.org/10.1541/ieejsmas.142.266

[2] Takafumi Yamaguchi, Naoto Usami, Kei Misumi, Atsushi Toyokura, Akio Higo, Shimpei Ono, Gilgueng Hwang, Guilhem Larrieu, Yoshiho Ikeuchi, Agnès Tixier-Mita, Ken Saito, Timothée Lévi and Yoshio Mita, "Self-Deformable Flexible MEMS Tweezer Composed of Poly(Vinylidene Fluoride)/Ionic Liquid Gel for Electrical Measurements and Soft Gripping", Journal of Microelectromechanical Systems, Vol. 31(5), pp. 802-812, (October 2022). DOI: 10.1109/JMEMS.2022.3187428

Conference Presentations (selected)

[3] Agnès Tixier-Mita, Ihida Satoshi, Dongchen Zhu, Pierre-Marie Faure, Yasuyuki Sakai, Timothée Lévi, and Hiroshi Toshiyoshi, "Cell Culture Multimodal Electrical Analyses with Thin-Film-Transistor Sensing Platform", 128th annual meeting of the japanese association of anatomists (第128回日本解剖学会総会・全国学術集会), Sendai, Japan, March 18-20 2023. (Invited)

[4] Agnès Tixier-Mita, Satoshi Ihida and Hiroshi Toshiyoshi, “Bio-sensors based on Thin-Film-Transistor Technology”, The 70th JSAP Spring Meeting 2023, (2023 年第 70 回応用物理学会春季学術講演会), Tokyo, Japan, March 15-18 2023. (Invited)

[5] Kikuo Komori, Dongchen Zhu, Ayano Takenouchi, Yuma Hori, Satoshi Ihida, Yasuyuki Sakai, Hiroshi Toshiyoshi, Hiroshi Kimura and Agnès Tixier-Mita, “Towards the Development of Qualitative and Quantitative Analytical Devices for On-site Rapid Evaluation of Biomacromolecules in Cell-based Bioassays”, The 35th Annual Meeting of the Japanese Society for Alternatives to Animal Experiments (日本動物実験代替法学会 第 35 回大会) JSAAE'2022, Shizuoka, Japan, November 18-20 2022.

[6] Dongchen Zhu, Anne-Claire Eiler, Satoshi Ihida, Yasuyuki Sakai, Hiroshi Toshiyoshi, Agnès Tixier-Mita, Kikuo Komori, “In-vitro Electrophysiology Evaluation of Pancreatic Beta-cells based on Thin Film Transistor Microelectrode Array”, The 35th Annual Meeting of the Japanese Society for Alternatives to Animal Experiments (日本動物実験代替法学会 第 35 回大会) JSAAE'2022, Shizuoka, Japan, November 18-20 2022.

[7] Tieying Xu, Satoshi Ihida, Hiroshi Toshiyoshi and Agnès Tixier-Mita, “Instrumentation development for 2D bioimpedance mapping with a Thin-Film-Transistor active matrix device”, 2022 Symposium on Design, Test, Integration and Packaging of MEMS/MOEMS (DTIP'2022), Pont-a-Mousson, France, July 11-13 2022. DOI: 10.1109/DTIP56576.2022.9911743

[8] Agnès Tixier-Mita, Satoshi Ihida, Anne-Claire Eiler, Tieying Xu, Pierre-Marie Faure, Timothée Lévi and Hiroshi Toshiyoshi, “Thin-Film-Transistor Sensing Platform for Real-time Multi-modal Analyses of Excitable Cells Culture”, NEURO'2022, Okinawa, Japan, June 30 - July 3 2022. (Invited)

Instrumentation development for 2D bioimpedance mapping with a Thin-Film-Transistor active matrix device

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***Abstract*—Multimodal biosensor active matrix devices, integrated with electronic circuits, are tremendously attractive for biomedical application. Actually, with these devices, multiple dye-free sensing techniques can be applied until single cell level, for real-time measurement and improved efficiency. Besides CMOS technology, the realization of these devices can be achieved with Thin-Film-Transistor (TFT) technology, with the advantage of transparency and 10 times larger scale sensing area. In this abstract, the authors present the instrumentation development to achieve 2D bioimpedance sensing with TFT-devices. First results are also shown. This development will be combined to electrophysiology measurements, already obtained previously, and optical observation to achieve a 2D multimodal sensing system to investigate cardiomyocyte cell culture from different aspects.**

Keywords—bioimpedance, thin-film-transistors, cell culture monitoring, 2D mapping

I. Introduction

In-vitro biological cell analyses is fundamental for all biological sciences to understand how cell work and communicate, how complex organs get built and operate, and for disease modeling. Besides microscopic optical techniques, usually used for analyses [1, 2], electrical techniques using biosensors integrated directly at the level of the cells propose advanced approaches for real-time, long-term, high throughput and complementary analyses [3].

Recently the development of multimodal biosensing devices, combining several sensing techniques on the same device, is becoming tremendously attractive to analyze cells with the same device, from different point of view. In particular CMOS multimodal sensing devices, with active matrix and resolution under single cell, are rapidly emerging [4]. Meanwhile, Thin-Film-Transistor technology, mainly used in the field of display technology, gives also active matrix devices, with the advantage of transparency of the substrates and the sensors for optical observation, as well as a large sensing area (cm size, compared to mm size for CMOS technology). These features are very interesting to study complex and large scale *in-vitro* multi-cell culture (like neuro-cardiac culture) with a resolution until single cell. Our group has already demonstrated various application with these devices: dielectrophoresis, electrophysiology, bioimpedance sensing and electrochemistry [5, 6].

Bioimpedance sensing is a technique used to characterize cells. For instance, by measuring the spatial variation of impedance, it is possible to distinguish areas with no cells, dead cells, healthy cells and so on. A concept figure of our target is shown on Fig. 1. With TFT-devices, we reported only 1D sensing along one line of sensors [7], because the precedent set-up did not allow sensing in 2D. In this abstract, we would like to present the instrumentation development thanks to which 2D bio-impedance sensing becomes possible. First results of 2D bio-impedance sensing on a small matrix, with 3 different environments (air, deionized water DIW and phosphate buffer saline medium PBS), are also shown.

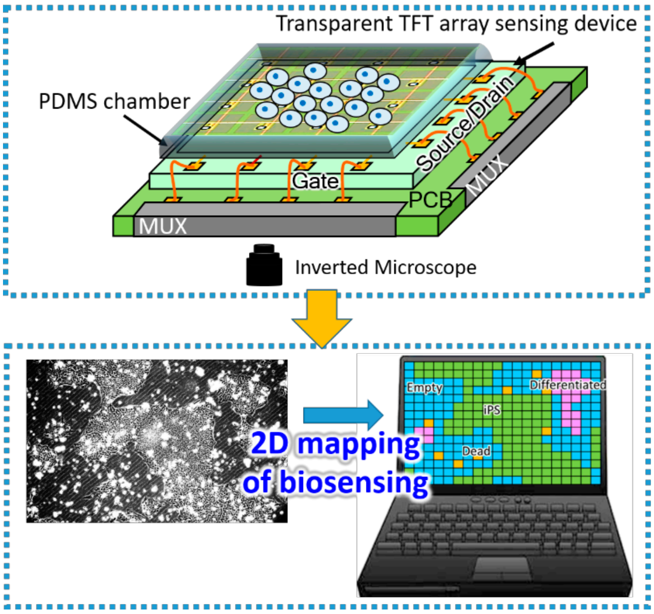

Fig. 1. Purpose of 2D bioimpedance mapping for cell culture monitoring. Thanks to the TFT matrix transparency, such electrical monitoring can be confirmed by optical observation at the same time.

Self-Deformable Flexible MEMS Tweezer Composed of Poly(Vinylidene Fluoride)/Ionic Liquid Gel for Electrical Measurements and Soft Gripping

Takafumi Yamaguchi, *Graduate Student Member, IEEE*, Naoto Usami, *Member, IEEE*, Kei Misumi, *Graduate Student Member, IEEE*, Atsushi Toyokura, Akio Higo, *Member, IEEE*, Shimpei Ono, Gilgueng Hwang, *Member, IEEE*, Guilhem Larrieu, *Member, IEEE*, Yoshiho Ikeuchi, Agnés Tixier-Mita, *Member, IEEE*, Ken Saito, *Member, IEEE*, Timothée Lévi, *Member, IEEE*, and Yoshio Mita, *Senior Member, IEEE*

***Abstract*— We present a self-deformable flexible tweezer capable of simultaneous mechanical handling and electrical measurements. The tweezer has a soft cantilever with the dimensions 2 mm × 8 mm × 75-100 μm, and undergoes self-deformation. The device is shown to be successfully capable of detecting electrical signals by gently touching the surface and grasping a spherical bead. The device demonstrated the lowest working voltage (1.5 V_{DC}), force suitable for soft gripping, and curvature radius of 2 mm, that was one of the smallest values compared to that of similar state-of-the-art devices. The device was fabricated using a unique and highly reliable process that was specifically developed to produce flexible cantilevers with novel ionic polymer-metal composites (IPMCs). The materials used were poly(vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE) and an ionic liquid (IL). The PVDF-TrFE/IL gel was prepared using acetone as the solvent and the gel was coated with silver nanowires as the electrodes. The actuator with a length of 8 mm and containing 50 wt% IL yielded the largest bending displacement of 7 mm and minimum curvature radius of 2 mm at 1.5 V_{DC}. [2022-0026]**

***Index Terms*— Electrical detection, flexible actuator, ionic liquid, PVDF-TrFE, self-deformable cantilever, silver nanowire, soft gripping.**

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This article has supplementary material provided by the authors and color versions of one or more figures available at https://doi.org/10.1109/JMEMS.2022.3187428.

Digital Object Identifier 10.1109/JMEMS.2022.3187428

I. Introduction

Microgrippers are expected to be the next-generation devices for various applications such as manipulation, assembly, or pick-and-place tasks by grabbing the microobjects [1]. Different methods of their actuations have been studied such as piezoelectric [2]–[4], electrostatic [5], [6], electrothermal [4], [7], [8] and ionic polymer-metal composite (IPMC) [9]–[12] actuators. Jain *et al.* (2014) [2] designed the three degree-of-freedom-based microgripper with piezoelectric actuator for microassembly. It involved conducting sequential pick-and-place of a micro pin in a hole driven by robust control of the applied voltage. Jaiswal *et al.* (2017) [3] fabricated the piezoelectric microtweezer with the flexible SU-8 end effector to measure the stiffness of spherical cells. Its force sensing by optical observation of the deflection of the actuator enables wide measurement of the forces ranging from less than one hundred nN to one mN. The main advantages of the piezoelectric grippers are small displacement

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IEEJ Transactions on Sensors and Micromachines
Vol.142 No.10 pp.266-272 DOI: 10.1541/ieejsmas.142.266

Paper

Real-time High-resolution Measurement of Pancreatic β Cell Electrophysiology Based on Transparent Thin-film Transistor Microelectrode Arrays

Dongchen Zhu[*a)] Member, Anne-Claire Eiler[**] Non-member
Satoshi Ihida[**] Non-member, Yasuyuki Sakai[*] Non-member
Hiroshi Toshiyoshi[**] Senior Member Agnès Tixier-Mita[**] Member
Kikuo Komori[*,***] Non-member

(Manuscript received Nov. 1, 2021, revised May 30, 2022)

A transparent high density thin-film-transistor microelectrode array (TFT-μEA) was investigated, for the first time, to apply to real-time electrophysiological monitoring on glucose-stimulated insulin secretion dynamics of pancreatic β cells at higher resolution than conventional microelectrode arrays (MEAs). TFT-μEAs employed in this work are designed based on the switch matrix architecture, which incorporates a large sensing area (15.6 mm × 15.6 mm) with a 150 × 150 array of indium-tin-oxide (ITO) microelectrodes placed at a 100 μm pixel pitch. TFT-μEAs coated with poly-L-lysine and laminin enabled to culture rat insulinoma β line iGL for at least 7 days without cell death, which was determined by conventional cell viability tests based on a fluorescent staining method. Real-time action potentials of iGL cells stimulated by 15 mM glucose were successfully observed in similar to those in a conventional MEAs. These results are the first step towards the development of a multimodal TFT-μEAs device for electrophysiological, biochemical and optical analyses of the pancreatic islets. TFT-μEAs would extremely be promising platforms in the bioanalysis field for neurochemistry and electrophysiology.

Keywords : thin film transistor, microelectrode array, pancreatic β cell

1. Introduction

As the functional unit of the pancreas, islets of Langerhans play vital roles in regulating glycaemia by secreting insulin at the right mom ent and in appropriate amounts. Dysfunction of the insulin-secreting β cells perturbs glucose homeostasis, leading to life-threatening hypoglycemia due to excessive insulin secretion or to damaging hyperglycemia owing to insufficient insulin secretion[(1)(2)]. To elucidate the pathophysiology of islet-related diseases, new approaches are needed to study islets at the cellular level.

The electrophysiology of pancreatic β cells has been investigated using a patch-clamp technique[(3)]. Kanno et.al have investigated the regulation of K^+ current in the presence of 10 mM glucose by applying perforated patch-clamp to mouse pancreatic islets[(4)]. However, patch-clamp technique is invasive to cells with complex operation. Moreover, it is difficult to record long-term electrophysiology of pancreatic β cells for hours using patch clamp technique[(5)]. Microelectrode array (MEA), on the other hand, have been employed to record electrical activities of islets during insulin secretion[(6)]. Pfeiffer et.al have detected burst and inter-burst phase of membrane potential oscillations using MEAs and correlated length of burst phase with the amount of insulin release[(7)]. Owing to low density and numbers of electrodes on a limited active area (0.05 mm^2), detailed two-dimensional analysis of tissues with larger diameter using MEAs is difficult. To overcome this issue, development of a two-dimensional high-resolution visualization system for bioanalytical image applications is required.

Thin-film-transistor microelectrode arrays (TFT-μEAs) as a microelectromechanical system originating from flat panel display technology have attracted great attention as one of the powerful chemical and bioanalytical tools thanks to their large surface area, higher electrode density and optical transparency. As shown in Table 1, the electrode density of the conventional MEAs is only around 1.3% owing to their small electrodes and relatively large electrode gaps[(7)], meanwhile the TFT-μEAs exhibit considerably larger electrode density (81.0%～85.5%). Therefore, monitoring electrophysiology of nearby or conjugated cells is particularly expected to be much more accessible using the high-resolution TFT-μEAs, compared to the conventional MEAs. Complementary metal oxide semiconductor (CMOS)-MEAs, on the other hand, have relatively larger active area (～8.09 mm^2) with thousands of high-resolution electrodes[(8)]. However, biological studies using optical microscope is much more difficult on non-transparent CMOS-MEAs when compared to TFT-μEAs.

Recently, Tixier-Mita et al. have successfully detected the presence of living yeast cells on the TFT-μEAs by measuring impedance variation and microphotographs simultaneously[(9)]. Cathcart et al. have conducted a two-dimensional mapping analysis

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*** Department of Biotechnology and Chemistry, Kindai University Takaya-Umenobe, Higashi-Hiroshima 739-2116, Japan

TOCHIGI Laboratory

Current Research Activities 2022-2023

Tochigi Laboratory

1. Research Topics

1.1 Investigation of the behavior of lattice defects by in situ mechanical testing

The mechanical properties of crystalline materials are governed by the behavior of lattice defects, such as dislocations, twins, and interfaces. We investigate the mechanical response of lattice defects by in situ loading experiments with a transmission electron microscope (TEM). A custom-made device fabricated by microelectromechanical system (MEMS) technology is employed as a loading mechanism. This device provides precise load control of sub-micronewton for a nano-sized test piece. The test pieces are prepared by focused ion beam (FIB), their shape has some manufacturing fluctuation. A detailed shape evaluation is required for quantitative mechanical experiments. Fig. 1 shows a scanning TEM (STEM) image of a typical test piece and corresponding thickness map evaluated by electron energy loss spectroscopy (EELS), indicating that the test piece has a thickness of approximately 50 nm. This kind of information on the shape evaluated by the present method is indispensable to estimate local stress during in situ loading experiments.

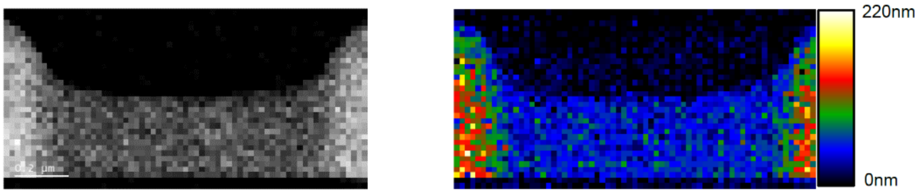

Fig. 1. STEM image and thickness map of a typical test piece for in situ mechanical testing.

Fig. 2 shows an atomic-resolution STEM image of a dislocation introduced in $SrTiO_3$ by in situ loading experiments. The bright spots correspond to atomic columns, as indicated by the structure model at the bottom right. Systematic loading and unloading experiments demonstrated that the motion of the dislocation core along the vertical direction. This implies that the dislocation migrates by the climb mechanism with atomic diffusion. We will further analyze the experimental data obtained and examine the relationship between dislocation motion and atomic structure transition.

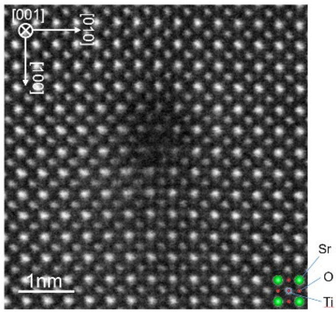

Fig. 2. Atomic-resolution STEM image of dislocation in $SrTiO_3$.

1.2 Development of in situ experimental systems

We developed new in situ experimental systems to expand our experimental capability. As mentioned in the previous section, in situ TEM technique is useful to examine nano- to atomic-scale phenomena. However, this is a relatively low throughput method, and the size of test pieces is limited. To overcome such problems, a loading MEMS device holder compatible with scanning electron microscopes (SEM) has been developed (Fig. 3). This apparatus can conduct ex-ante evaluations, such as performance test of MEMS device and pre-loading test of samples just after sample preparation, which highly improves the throughput of our experiment works.

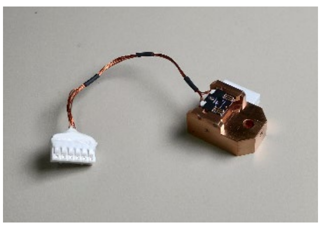

Fig. 3. MEMS device holder for in situ SEM mechanical testing.

2. Research Achievements

2.1 Number of original journal paper: 1

2.2 International conferences: 3 (including 2 invited presentations)

2.3 Domestic conferences: 5

2.4 Number of patents: 0

3. List of awards

Not available

4. Research Grants

4.1 Total number of research grants: 7

4.2 Number of collaboration research with industries: 0

4.3 List of major research grants (serving as Principal Investigator)

- Grant-in-Aid for Scientific Research B "*Investigation of atomistic mechanisms of twinning based on direct observations*" from JSPS.
- PREST "Atomic-scale investigations of deformation and fracture phenomena" from JST.

5. Education

5.1 Number of Ph.D. students (including current students): 0

5.2 Number of master students (including current students): 0

5.3 Number of other students: 0

6. Publication list

Journal Papers

1. Yan Li, Xufei Fang, Eita Tochigi, Yu Oshima, Sena Hoshino, Takazumi Tanaka, Hiroto Oguri, Shigenobu Ogata, Yuichi Ikuhara, Katsuyuki Matsunaga, Atsutomo Nakamura, "Shedding new light on the dislocation-mediated plasticity in wurtzite ZnO single crystals by photoindentation", Journal of Materials Science & Technology, **156** (2023) 206-216.

International Conference Presentations

1. Eita Tochigi, "Atomic-scale investigations of deformation and fracture behavior of ceramic materials" CIMTEC 2022, Perugia, Italy (2022).
2. Eita Tochigi, "Investigation of local mechanical responses in ceramic materials based on in situ TEM observations" MS&T 2022, Pittsburgh, USA (2022).
3. Eita Tochigi, Takaaki Sato, Minjian Cao, Naoya Shibata, Hiroyuki Fujita, Yuichi Ikuhara, " Analysis of mechanical behavior of crystalline materials by atomic-resolution in situ TEM loading experiment", ICMR 2022, Yamaguchi, Japan (2022).

Symposium CC
Modelling, Simulation and Testing of Mechanical and Thermomechanical Properties of Bulk Ceramics, Coatings and Composites

ABSTRACTS

CC-1:IL11 **Atomic-scale Investigations of Deformation Behaviour of Ceramic Materials**

EITA TOCHIGI, Institute of Industrial Science, The University of Tokyo, Tokyo, Japan

Deformation of crystalline materials is attributed to atomic displacements induced by mechanical stress. To understand the deformation behavior in detail, atomic scale analysis is essential. In this study, we investigate the mechanical responses of oxide crystals from nano to atomic scale. Firstly, the behavior of rhombohedral twinning in sapphire (α-Al2O3) is discussed. By in situ transmission electron microscopy (TEM) nanoindentation, we show that twinning/detwinning phenomena occurred by the glide motion of twinning dislocations on the matrix/twin interfaces. Furthermore, the atomic motions associated with the twinning dislocation glide is demonstrated by atomic-resolution scanning TEM and first-principles molecular dynamics simulation. It is revealed that the elementary process of the twinning corresponds to cooperative motions of a grope of five atoms. Secondly, we introduce a custom-made loading device for in situ TEM mechanical experiment. The loading device was fabricated by micro electro mechanical system (MEMS) technique, and it has a good load resolution of sub-μN. We show some results of local strain changes in SrTiO3 upon loading measured based on atomic-resolution scanning TEM images.

Analysis of mechanical behavior of crystalline materials by atomic-resolution in situ TEM loading experiment

E. Tochigi[1,2*], T. Sato[1], M. Cao[3], N. Shibata[3,4], H. Fujita[5], Y. Ikuhara[3,4]

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1. Introduction

Plastic deformation of crystalline materials is attributed to atomic displacements. Therefore, it is essential to understand atomic structure evolution upon loading. In situ transmission electron microscopy (TEM) mechanical testing is a powerful technique to directly observe the microstructural changes. However, in situ TEM sample holders for mechanical testing are relatively large and thus not typically compatible with atomic-resolution TEMs having a small sample chamber. In recent years, micromachining techniques have remarkably advanced, and mechanical devices in micrometer scale, i.e., microelectromechanical systems (MEMS), have been put into practical use. MEMS devices are produced from a Si-based substrate with flexible design and can be equipped with a precise loading mechanism within micrometer size. Therefore, MEMS devices can be a promising experimental system for in situ TEM mechanical testing. Indeed, there are several reports on in situ mechanical testing using MEMS devices [1-3], although it is still difficult to perform the mechanical testing with atomic-resolution imaging. In this study, we performed atomic-resolution in situ mechanical testing using a custom-made MEMS loading device in a TEM and examined local strain distributions of crystalline materials upon loading.

2. Experimental procedure

$SrTiO_3$ single crystal was selected as a test material in this study. A $SrTiO_3$ substrate was cut into a small piece, and it was fixed on a MEMS loading device by focused ion beam (Helios G4, Thermo Fisher Scientific). The sample was further milled to a pair of L-shaped parts. A notch was made on one of the L-shaped parts and its edge was thinned down for electron transparency. The MEMS loading device was controlled through a double tilt biasing holder (Aduro 300, Protochips) and a source measure unit in an atomic-resolution scanning TEM (STEM: ARM-200F, JEOL).

3. Results and discussion

Figure 1 shows a custom-made MEMS loading device for in situ TEM mechanical testing. This device has two beams driven by the electrostatic actuators. A sample was fixed at the cross points of the arms. The force applied to the sample can be calculated from the driving voltage, the displacement of the arm and its stiffness. Detailed mechanisms of this loading device are given elsewhere [3].

Figure 2 shows a high-angle annular dark-field (HAADF) image of $SrTiO_3$ sample taken during in situ experiment at an applied load of 13.3 μN. It is found that sphere contrasts corresponding to atomic columns are clearly visualized upon loading, where the strong and weak white contrasts correspond to Sr and Ti-O atomic columns respectively. By comparison to the rectangular shape drawn as guidelines, the atomic lattice is found to be distorted due to the tensile force applied along the [110] directions.

To measure local strain distribution, each atomic column position was precisely identified from the HAADF image by gaussian fitting approach using a following function:

$$f(x,y) = Aexp\left[-\frac{(x-x_0)^2}{2\sigma_x^2}\right]exp\left[-\frac{(y-y_0)^2}{2\sigma_y^2}\right] + C \quad (1).$$

The six fitting parameters were estimated by the Markov chain Monte Carlo method using the random-walk Metropolis-Hastings algorithm [4]. The strain distributions (ε_{xx}) corresponding to the HAADF image of Figure 2 are shown in Figure 3. Tensile strain appears over almost all the area, and relatively strong strain distributions spread out from the notch due to stress concentration there.

The present results showed that our in situ TEM loading system can visualize the mechanical response of crystalline materials at the atomic level. This technique provides us a novel means of exploring nanomechanical behavior.

Summary

In situ TEM loading testing was carried out for $SrTiO_3$ single crystal. Atomic-resolution HAADF images of the sample upon loading were

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Research Article

Shedding new light on the dislocation-mediated plasticity in wurtzite ZnO single crystals by photoindentation

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A B S T R A C T

Dislocation-mediated plasticity in inorganic semiconductors and oxides has attracted increasing research interest because of the promising mechanical and functional properties tuned by dislocations. In this study, we investigated the effects of light illumination on the dislocation-mediated plasticity in hexagonal wurtzite ZnO, a representative third-generation semiconductor material. A (0001) 45° off sample was specially designed to preferentially activate the basal slip on (0001) plane. Three types of nanoindentation tests were performed under four different light conditions (550 nm, 334 nm, 405 nm, and darkness), including low-load (60 μN) pop-in tests, high-load (500 μN) nanoindentation tests, and nanoindentation creep tests. The maximum shear stresses at pop-in were found to approximate the theoretical shear strength regardless of the light conditions. The activation volume at pop-ins was calculated to be larger in light than in darkness. Cross-sectional transmission electron microscope images taken from beneath the indentation imprints showed that all indentation-induced dislocations were located beneath the indentation imprint in a thin-plate shape along one basal slip plane. These indentation-induced dislocations could spread much deeper in darkness than in light, revealing the suppressive effect of light on dislocation behavior. An analytical model was adopted to estimate the elastoplastic stress field beneath the indenter. It was found that dislocation glide ceased at a higher stress level in light, indicating the increase in the Peierls barrier under light illumination. Furthermore, nanoindentation creep tests showed the suppression of both indentation depth and creep rate by light. Nanoindentation creep also yielded a larger activation volume in light than in darkness.

1. Introduction

In recent decades, inorganic semiconductors have attracted enormous attention owing to their diverse applications in integrated circuits, optoelectronics, catalysis, and power electronics [1–5]. Research has focused on their distinct structural, optical, electrical, and optoelectronic properties [2,6], while their mechanical properties have received less attention. Conventional knowledge suggests that the majority of inorganic semiconductors are brittle, hard, and exhibit little or almost no dislocation-mediated plasticity at room temperature [7,8]. This lack of intrinsic plasticity comes from the intrinsic material features of chemical bonding and defects, such as strong ionic or directional covalent bonds between constituent atoms and poor dislocation mobility. Therefore, plastic deformability in inorganic semiconductors is generally achieved by heating samples to a high temperature to take advantage of thermal activation [9] or reducing the sample dimensions to submicron or nanoscale levels [10,11]. Bulk plasticity under ambient conditions remains a great challenge and a long-sought objective for inorganic semiconductors.

Recent research has opened up new prospects for plastically deformable inorganic semiconductors [12–15]. Shi et al. reported

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Engineering Ceramics: Microstructure-Property-Performance Relations and Applications: Processing-Microstructure-Property Relations of Engineering Ceramics
Sponsored by: ACerS Engineering Ceramics Division
Program Organizers: Young-Wook Kim, University of Seoul; Hua-Tay Lin, Guangdong University of Technology; Junichi Tatami, Yokohama National University; Michael Halbig, NASA Glenn Research Center

Wednesday 8:00 AM
October 12, 2022
Room: 415
Location: David L. Lawrence Convention Center

Session Chair: Yiquan Wu, Alfred University; Soshu Kirihara, Osaka University

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8:00 AM Invited

Investigation of Local Mechanical Responses in Ceramic Materials Based on In Situ TEM Observations: *Eita Tochigi*[1]; [1]The University of Tokyo

Plastic deformation and fracture of ceramic materials typically initiate from a stress concentration region at a surface crack. Therefore, it is important to characterize the microscopic behavior of a surface crack under loading conditions. In situ transmission electron microscopy (TEM) mechanical experiments are known to be a powerful technique to visualize local structural changes upon loading. In recent years, microelectron mechanical system (MEMS) technology provides small and precise loading devices. In this study, we performed in situ loading experiments on ceramic materials with a MEMS loading device in an atomic-resolution scanning transmission electron microscope. Our experiments provided sequential atomic-resolution images. The local strain of the sample was examined based on atomic positions, and the results clearly show increasing of local strain with increasing load and following deformation/fracture behavior. In the presentation, we will discuss its strain distributions and structural changes in detail.

MATSUHISA Laboratory

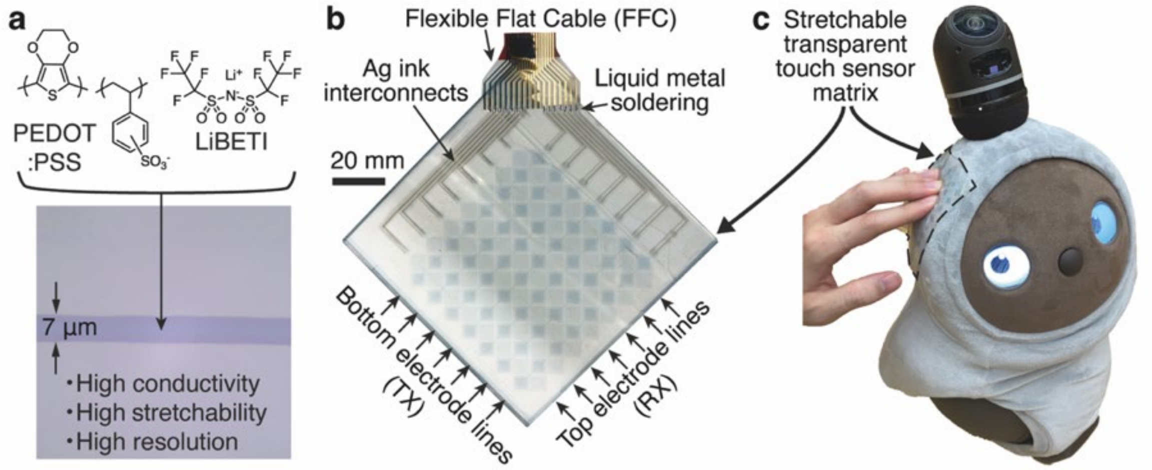
Current Research Activities 2022-2023

Matsuhisa Laboratory

1. Research Topics

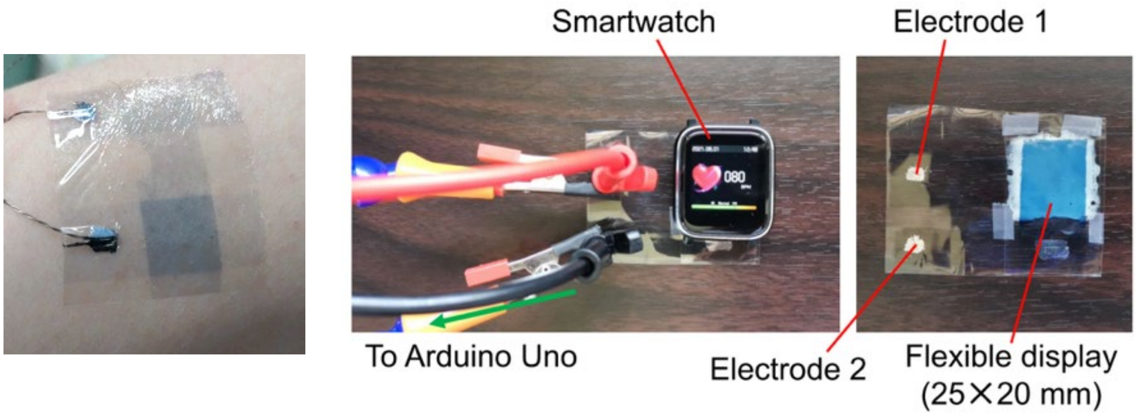
1.1 A high-resolution, transparent, and stretchable polymer conductor for wearable sensor arrays

We have enabled a polymer conductor with high conductivity, stretchability, high-resolution patternability, and optical transparency (Figure a) [1]. In the previous design of stretchable conducting polymers, it was difficult to design materials that simultaneously maintained these properties. Our conducting polymer was realized by decoupling the engineering of the materials properties. High conductivity was achieved using a rationally designed additive (LiBETI). High stretchability was achieved by matching the mechanical properties with those of the substrate. To the best of our knowledge, a crack-on-set strain of 160% has the highest value among previous reports. The developed conducting polymer was patterned at a resolution of less than 10 µm using nanosecond UV laser ablation. Moreover, we demonstrated two applications to show the high feasibility of our polymer conductors. The first is a stretchable transparent touch sensor matrix that can be easily implemented on arbitrary surfaces, including soft robotic skins (Figure b and c). The second is a high-resolution strain sensor that can map pulse-wave signals. Our findings can further accelerate the design of high-performance stretchable conducting polymers and their future applications in next-generation wearable devices or robotic skins.

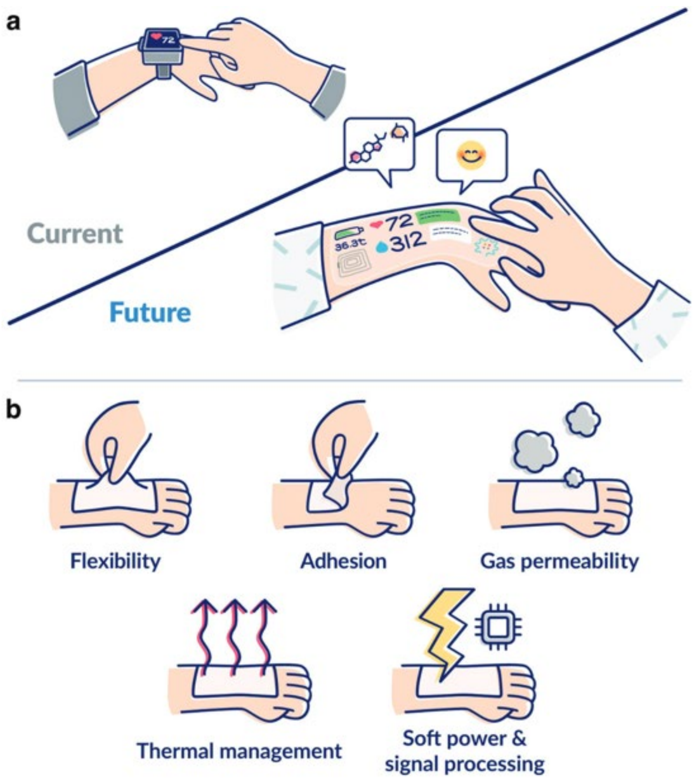


1.2 A stretchable electrochromic display with exceptional skin conformability

Skin-like stretchable electronic devices are expected to lead to the next generation of advanced healthcare and health monitoring. Unlike conventional rigid devices, the development of soft devices with high skin-conformability is required. Displays that adhere to the skin can show a variety of information as the next-generation wearable devices. Among the stretchable displays, electrochromic displays (ECDs) can be driven by low voltage and are highly stable in air. However, ECDs reported so far are thick (about 50 µm),1 which are difficult to show high skin-conformability (Figure Left). Stretchable displays that adhere to the skin and operate stably in air are required. In this study, we demonstrate an ultrathin (<10 µm) stretchable ECD with high skin-conformability. Our ultrathin and stretchable ECDs worked stably even under 50% tensile strain, which is sufficient to allow ECDs to adhere to the skin and operate. Furthermore, our ECDs worked with a line width and spacing of 50 µm. High-resolution displays were realized. In the future, it is expected that the biological signals information read by the sensor can be displayed directly on the skin by a skin display. Furtheremore, our ECD succeeded in producing pulse wave signals for prosthetic hands (Figure Right) [2].



1.3 <u>Engineering The Comfort-of-Wear For Next Generation Wearables (Review)</u>
We gave a focus on the "comfort-of-wear" of future wearables. Wearable technologies are becoming important for information technology and healthcare [7]. The demand on society is increasing because of ageing societies, chronic diseases, and pandemic diseases. Recently developed flexible/stretchable devices have shown their potential in long-term healthcare monitoring with improved signal integrity and multimodality. However, the actual users' adherence to such wearable devices cannot be determined not only by the function. We identify "comfort-of-wear" as one of the most critical parameters for future wearables, similar to how we choose daily clothes as a matter of comfort. "Comfort-of-wear" is defined as the ability of the wearable to not disturb the wearers' daily life. We provide the reader with the analysis of several engineering approaches that can be used to improve the wear-of-comfort of wearable devices. Examples of outlined strategies include: reducing bending stiffness by structural and materials approaches, using breathable materials, and reducing the use of rigid components by system design.



2. Research Achievements

2.1 Number of original journal papers: 8

2.2 International conference: 19 (including 17 invited presentations)

2.3 Domestic conference: 12

2.4 Number of patents: 0

3. List of awards

- Telecommunications Systems Technology Award, "High-frequency and intrinsically stretchable polymer diodes" March 23rd 2023, Telecommunications Advancement Foundation (N. Matsuhisa, S. Niu *et al.*)
- MIT Technology Review Innovators Under 35 Global, June 29th 2022, MIT Technology Review (N. Matsuhisa)
- Young Scientist Presentation Award, June 17th 2022, The Japan Society of Applied Physics (N. Matsuhisa)
- Light upon the mountain award, April 13th 2022 (N. Matsuhisa)

4. Research Grants

4.1 Total number of research grants: 5

4.2 Number of collaboration research with industries: 2

4.3 List of major research grants (serving as Principal Investigator)

- PRESTO "Ultraflexible diodes using stretchable conductors and semiconductors" from JST
- Grant-in-Aid for Scientific Research B "Mimicking skin sensory organs by multi-modal ultraflexible electronic materials" from JSPS

5. Education

5.1 Number of Ph.D. students (including current students): 0

5.2 Number of master students (including current students): 3

5.3 Number of other students: 4

6. Publication list

Journal Papers

1. T. Shimura, S. Sato, T. Tominaga, S. Abe, K. Yamashita, M. Ashizawa, T. Kato, H. Ishikuro, N. Matsuhisa* "A High‐Resolution, Transparent, and Stretchable Polymer Conductor for Wearable Sensor Arrays" Advanced Materials Technologies, 2201992 (2023).
2. A. Fujii, K. Murao*, N. Matsuhisa "Pulse Wave Generation Method for PPG by Using Display" *IEEE Access* **11,** 31199-31211 (2023).
3. Y. Luo, M. R. Abidian, J.-H. Ahn, D. Akinwande, A. M. Andrews, M. Antonietti, Z. Bao, M. Berggren, C. A. Berkey, C. J. Bettinger, J. Chen, P. Chen, W. Cheng, X. Cheng, S.-J. Choi, A.

Chortos, C. Dagdeviren, R. H. Dauskardt, C.-A. Di, M. D. Dickey, X. Duan, A. Facchetti, Z. Fan, Y. Fang, J. Feng, X. Feng, H. Gao, W. Gao, X. Gong, C. F. Guo, X. Guo, M. C. Hartel, Z. He, J. S. Ho, Y. Hu, Q. Huang, Y. Huang, F. Huo, M. M. Hussain, A. Javey, U. Jeong, C. Jiang, X. Jiang, J. Kang, D. Karnaushenko, A. Khademhosseini, D.-H. Kim, I.-D. Kim, D. Kireev, L. Kong, C. Lee, N.-E. Lee, P. S. Lee, T.-W. Lee, F. Li, J. Li, C. Liang, C. T. Lim, Y. Lin, D. J. Lipomi, J. Liu, K. Liu, N. Liu, R. Liu, Y. Liu, Y. Liu, Z. Liu, Z. Liu, X. J. Loh, N. Lu, Z. Lv, S. Magdassi, G. G. Malliaras, N. Matsuhisa, A. Nathan, S. Niu, J. Pan, C. Pang, Q. Pei, H. Peng, D. Qi, H. Ren, J. A. Rogers, A. Rowe, O. G. Schmidt, T. Sekitani, D.-G. Seo, G. Shen, X. Sheng, Q. Shi, T. Someya, Y. Song, E. Stavrinidou, M. Su, X. Sun, K. Takei, X.-M. Tao, B. C. K. Tee, A. V.-Y. Thean, T. Q. Trung, C. Wan, H. Wang, J. Wang, M. Wang, S. Wang, T. Wang, Z. L. Wang, P. S. Weiss, H. Wen, S. Xu, T. Xu, H. Yan, X. Yan, H. Yang, L. Yang, S. Yang, L. Yin, C. Yu, G. Yu, J. Yu, S.-H. Yu, X. Yu, E. Zamburg, H. Zhang, X. Zhang, X. Zhang, X. Zhang, Y. Zhang, Y. Zhang, S. Zhao, X. Zhao, Y. Zheng, Y.-Q. Zheng, Z. Zheng, T. Zhou, B. Zhu, M. Zhu, R. Zhu, Y. Zhu, Y. Zhu, G. Zou, X. Chen* "Technology Roadmap for Flexible Sensors" *ACS Nano* **17,** 5211-5295 (2023).
4. T. Nishikawa, H. Yamane, N. Matsuhisa, N. Miki* "Stretchable Strain Sensor with Small but Sufficient Adhesion to Skin" *Sensors* **23,** 1774 (2023).
5. J. Kang, J. Mun, Y. Zheng, M. Koizumi, N. Matsuhisa, H.-C. Wu, S. Chen, J. B.-H. Tok, G. H. Lee, L. Jin*, Z. Bao* "Tough-interface-enabled stretchable electronics using non-stretchable polymer semiconductors and conductors" *Nature Nanotechnology* **17,** 1265-1271 (2022).
6. Y. Kim, C. Zhu, W.-Y. Lee, A. Smith, H. Ma, X. Li, D. Son, N. Matsuhisa, J. Kim, W.-G. Bae, S. H. Cho, M.-G. Kim, T. Kurosawa, T. Katsumata, J. W.F. To, J. Y. Oh, S. Paik, S. J. Kim, L. Jin, F. Yan, J. B.-H. Tok, Z. Bao* "A Hemispherical Image Sensor Array Fabricated with Organic Photo-memory Transistors" *Advanced Materials* **35,** 2203541 (2023).
7. T. Shimura, S. Sato, P. Zalar*, N. Matsuhisa* "Engineering the Comfort‐of‐Wear for Next Generation Wearables" *Advanced Electronic Materials*, 2200512 (2022).
8. A. Abramson, C. T. Chan, Y. Khan, A. Mermin-Bunnell, N. Matsuhisa, R. Fong, R. Shad, W. Hiesinger, P. Mallick, S. S. Gambhir, Z. Bao* "A flexible electronic strain sensor for the real-time monitoring of tumor regression" *Science Advances* **8,** eabn6550 (2022).

Conference Presentations (selected)

[9] N. Matsuhisa “Rubber-like stretchable electronics for skin-conformable wearable devices” 2022 IEEE CPMT Symposium Kyoto, Japan, November 10th 2022.
[10] T. Tominaga, T. Shimura, M. Ashizawa, N. Matsuhisa “An ultrathin and stretchable electrochromic display with exceptional skin conformability” 13th International Conference on Nano-Molecular Electronics (ICNME2022), Tokyo, Japan, December 14th 2022.
[11] N. Matsuhisa " Soft and Highly Deformable Electronic Devices for Future Wearables" Electron Devices Technology and Manufacturing Conference (IEEE EDTM 2023), Seoul, Korea, March 9th 2023.
[12] N. Matsuhisa, "Rubber-like stretchable electronics for skin-conformable wearable devices" 2022 IEEE CPMT Symposium Japan, Kyoto, Japan, November 10th 2022.
[13] N. Matsuhisa, “Developing Smart Skin Devices” MIT Technology Review EmTech, Boston, USA, November 3rd 2022.
[14] N. Matsuhisa, “Intrinsically stretchable electronic materials for high-frequency, skin-conformable wearable devices” International Conference on Flexible and Printed Electronics 2022, Jeju, Korea, October 13th 2022.
[15] N. Matsuhisa, “Intrinsically stretchable electronic materials and devices” 2022 Symposium on VLSI Technology and Circuits, Hawaii, USA, June 14th 2022.

PHYSICAL SCIENCES

A flexible electronic strain sensor for the real-time monitoring of tumor regression

Alex Abramson[1]†, Carmel T. Chan[2,3], Yasser Khan[1]‡, Alana Mermin-Bunnell[1,4], Naoji Matsuhisa[1]§, Robyn Fong[5], Rohan Shad[5], William Hiesinger[5], Parag Mallick[2,6], Sanjiv Sam Gambhir[2,3,4,6,7]||, Zhenan Bao[1]*

Assessing the efficacy of cancer therapeutics in mouse models is a critical step in treatment development. However, low-resolution measurement tools and small sample sizes make determining drug efficacy in vivo a difficult and time-intensive task. Here, we present a commercially scalable wearable electronic strain sensor that automates the in vivo testing of cancer therapeutics by continuously monitoring the micrometer-scale progression or regression of subcutaneously implanted tumors at the minute time scale. In two in vivo cancer mouse models, our sensor discerned differences in tumor volume dynamics between drug- and vehicle-treated tumors within 5 hours following therapy initiation. These short-term regression measurements were validated through histology, and caliper and bioluminescence measurements taken over weeklong treatment periods demonstrated the correlation with longer-term treatment response. We anticipate that real-time tumor regression datasets could help expedite and automate the process of screening cancer therapies in vivo.

INTRODUCTION

In the process of clinical translation, thousands of potential cancer drugs are tested for every one drug that makes it to patients. Oncology researchers use a suite of in vitro high-throughput screening models that implement computational algorithms, genomics testing, cell culture, and organoid systems to assess the efficacy of these numerous drugs quickly and inexpensively against a given cancer type (*1*–*4*). In vivo models, however, generally produce results that more closely resemble clinical outcomes (*5*). Researchers typically read out in vivo models by comparing tumor volume regression between multiple replicates of treated and untreated controls. However, inherent biological variations combined with low-resolution measurement tools and small sample sizes make determining drug efficacy in vivo a difficult, labor-intensive task (*6*). Accurately determining treatment response is critical to clinical translation, and tools automating in vivo tumor regression measurements could facilitate this process by gathering high-resolution continuous datasets in larger animal cohorts. Such advances in data quality and labor reduction could lead to automated high-throughput in vivo drug testing setups and more accurate experimental results.

Here, we present an elastomeric-electronic tumor volume sensor capable of autonomously reading out cancer treatment efficacy studies in vivo. Using advances in flexible electronic materials (*7*–*12*), we designed a conformal, wearable strain sensor that continuously measures, records, and broadcasts tumor volume changes occurring in subcutaneously implanted tumors on the minute time scale. The sensor's real-time dataset enables us to track the immediate pharmacodynamic response of a given drug by recording significant tumor shrinkage continuously. In two unique tumor models, our sensor was able to discern differences in tumor volume dynamics between drug- and vehicle-treated tumors within hours following therapy initiation in vivo. Short-term regression measurements in these models were validated by histology taken within hours following therapy initiation, and caliper and bioluminescence measurements over weeklong treatment periods demonstrated correlation with longer-term treatment response.

This sensor achieves three main advances over other common tumor measurement tools such as calipers, implantable pressure sensors, and imagers. First, because the sensor remains in place over the entire measurement period and takes measurements every 5 min, it is possible to generate a four-dimensional (4D), time-dependent dataset that eliminates the need for any guesswork on measurement timing. Imaging techniques such as computed tomography (CT) and bioluminescence are unable to achieve these same time resolutions over long measurement periods. This is due to the toxicity limitations associated with the necessary radiation and contrast dye in CT imaging; in addition, high-resource and cost constraints prevent imaging scale up to larger cohorts or more frequent sampling time points (*13*). Moreover, implantable pressure sensors require invasive procedures that compromise the mechanical integrity of the tumor, and they work best when measuring tumors encapsulated within a solid environment such as bone (*14*). Second, the strain sensor has the capability of precisely distinguishing size changes that are difficult to detect using caliper and bioluminescence imaging measurements. This is due to the errors associated with the physical measurement of soft tissue (*15*–*17*) and the positive but inexact correlation between bioluminescence readouts and tumor volume (*18*), respectively. Third, the sensor is entirely autonomous and noninvasive. Thus, using it reduces the costs and labor associated with performing measurements and enables direct data comparisons between operators. Consequently, it enables fast, inexpensive,

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RESEARCH ARTICLE

Pulse Wave Generation Method for PPG by Using Display

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ABSTRACT The extensive research on wearable devices has led to devices with various shapes and mounting locations. Wearable devices are often used to record the user's biometric information, and methods have been proposed to detect physical abnormalities from the acquired data. Among various kinds of biometric data, pulse data has been used in methods such as heart rate monitoring and emotion recognition. The most common type of pulse sensor uses photoplethysmography (PPG), which irradiates a green LED on the skin and measures pulse data from changes in the light reflected from blood vessels. PPG sensors have been implemented in commercially available wearable devices such as smartwatches. However, a PPG sensor requires blood flow for data acquisition, and when a smartwatch is worn on an artificial body part such as a prosthetic hand or a robotic arm, data cannot be acquired because there is no blood flow. In this study, we propose a method that enables a PPG sensor to measure arbitrary pulse data by using a display. If this method is successful, it will enable pulse data measured at the junction of a living limb and an artificial limb to be input to the display; then, a smartwatch attached to the artificial limb will read the same pulse data. In this paper, we focus on the heart rate and report the results of an experiment in which a target heart rate was input and the display was controlled accordingly to determine whether the target heart rate could be obtained by a smartwatch. We implemented a display drawing program and conducted the evaluation with five kinds of smartwatches and four kinds of displays. The results showed that the error between the target heart rate and the heart rate acquired by the smartwatch was within 3 beats per minute (bpm) in many cases when the target heart rate was set to 60–100 bpm. When the target heart rate was set to 40–55 and 105–200 bpm, the heart rate could also be input into the smartwatch with a small error under certain conditions. Moreover, when generated PPG data was imported into heart rate variability (HRV) analysis software, it was recognized as a pulse wave in the same way as real PPG data obtained from a person. We compared the heart rate, RR interval, and SDNN calculated from the real and generated PPG data, and we confirmed that the proposed method could generate stable PPG data. On the other hand, when the waveforms were compared, the generated PPG waveform differed greatly from the real PPG waveform, which indicated that the software could calculate the heart rate, RR interval, SDNN, and LF/HF ratio regardless of the waveform. This result suggests that the calculation of these parameters without verifying the waveform would be vulnerable to an attack with fake PPG data.

INDEX TERMS Pulse wave, heart rate, biometric information, PPG sensor, smartwatch, display.

I. INTRODUCTION

With the growing awareness of health management, wearable devices that record biometric information have become widely used. The recorded biometric information includes a variety of data, such as activity, respiration, body temperature, cardiac potential, blood pressure, gaze, pulse wave, and heart rate. The pulse sensor used to acquire the latter two kinds of data (i.e., the pulse wave and heart rate) irradiates the skin with LEDs that emit infrared light, red light,

The associate editor coordinating the review of this manuscript and approving it for publication was Larbi Boubchir.

A High-Resolution, Transparent, and Stretchable Polymer Conductor for Wearable Sensor Arrays

*Tokihiko Shimura, Shun Sato, Taizo Tominaga, Shuma Abe, Kaoru Yamashita, Minoru Ashizawa, Takeo Kato, Hiroki Ishikuro, and Naoji Matsuhisa**

Arrays of stretchable and transparent electronic sensors realize next-generation skin-conformable wearables and soft robotic skins, which require a high-resolution patternable stretchable conductor. However, the difficulty of simultaneously engineering desirable material properties (i.e., conductivity, stretchability, and patternability) has limited the development of such stretchable electronic materials. Herein, a high-resolution patternable, stretchable, and transparent conducting polymer by decoupled engineering of the material properties is shown. The high conductivity of the conducting polymer is achieved by rationally designing an ionic additive. The high stretchability is realized by matching the mechanical properties of the conducting polymer to the substrate. The developed conducting polymer is then patterned in a resolution less than 10 µm by nanosecond UV laser ablation, which enables the feasible demonstration of stretchable and transparent sensor arrays for touch and strain. The findings in this work will accelerate the development of high-density stretchable sensor arrays and stretchable semiconductor devices.

1. Introduction

Spatially distributed sensing on soft and deformable surfaces, including human skin, is essential for next-generation human–computer interfaces (HCIs) and wearable healthcare sensors. Increasing the number of inputs gives more freedom in HCIs. Multipoint biological signal sensing allows us to detect the propagation of signals and to easily find the place of interest. Sensor arrays based on a thin elastomeric sheets are capable of spatial sensing, which can exhibit mechanical properties similar to that of human skin: stretchability of >30%[1] and Young's Modulus of ≈1 MPa.[2] Ideally, the sensors should be transparent. The aesthetic impression to the user can determine the adherence of wearable devices, with transparent devices minimally influencing the original appearance. Furthermore, transparency allows us to integrate optical healthcare sensors to add multimodality[3] or displays for the indication of obtained signals from the sensors.[4]

For the realization of stretchable and transparent sensor arrays, it is important to develop high-resolution patternable, transparent, and stretchable conductors. Stretchable and transparent conductors have been realized using liquid metals,[5] carbon nanotubes,[6] silver nanowires,[7] and conducting polymers.[8] Among them, conducting polymers, including poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS), are attractive because of the low intrinsic modulus, transparency, and biocompatibility. Stretchable conducting polymers have been mainly realized by mixing nonstretchable PEDOT:PSS with surfactants,[9,10] ionic additives,[11] sorbitol,[12] or ductile polymers,[13–16] to simultaneously realize high stretchability and conductivity. The patterning of stretchable conducting polymers was performed by utilizing inkjet printing[11,17,18] and photolithography[15,16,19] to yield a resolution of 40 and 2 µm, respectively.

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Engineering the Comfort-of-Wear for Next Generation Wearables

Tokihiko Shimura, Shun Sato, Peter Zalar, and Naoji Matsuhisa**

Wearable technologies are becoming important for the fields of information technology and healthcare, driven mainly by societal issues such as the aging society and the current pandemic. Recently developed flexible/stretchable wearable devices have demonstrated their ability for long-term healthcare monitoring with improved signal integrity and multimodality. However, the adherence of wearers to such wearable devices cannot be determined only by the function. Here "comfort-of-wear" is identified as one of the most critical parameters for future wearables, similar to how clothes are chosen based on how comfortable they are. "Comfort-of-wear" is defined as the device's ability to not to disturb the wearers' daily life. Several engineering approaches are introduced to improve the comfort-of-wear of devices—via strategies that include improving flexibility by utilizing a combination of structures, materials, and systems. Finally, the future of wearables enabled by cutting-edge advanced electronic technologies is proposed.

1. Introduction

Wearable devices are becoming a normal part of everyday life. In addition to functions like notifying you of recent updates in your social network or your schedule or storage of payment information, their ability to monitor the health of its wearer is of importance especially under crises like a world-wide pandemic or the aging society. The rigid and compact body of commercialized wearables can monitor our heart rate or blood oxygen levels (SPO_2) by photoplethysmography (PPG) and our heart's electrical activity by electrocardiography (ECG). In addition, they can monitor body activities and state (exercise, sleep, body temperature, and stress levels) which are useful for the detection of irregularities in our bodily function.

Still, the skin contact area, which is the interface where physiological signals are obtained, is limited. Although conventional electronic materials are rigid, structural engineering approaches or soft electronic materials approaches have realized soft and conformable devices that can wrap over the complex shape of our body.[1,2] Among other things, imparting skin conformability to the device can significantly improve the signal integrity and reduce motion artifacts.[3,4] Improved mechanical flexibility enables highly sensitive sensors for physical parameters such as strain, pressure, and temperature.[5–8] Furthermore, fluid-based sensors have been realized including sweat-chemical sensors.[9–11] Moreover, skin-conformable displays have been realized to let wearers know the result of the physiological signals monitored by soft sensors.[12–15]

In addition to their sensing capability, it is very important to design for the "comfort-of-wear" of wearable devices. Comfort-of-wear can be defined by how the devices worn by the wearers can minimize the irregularity caused by the device's physical existence in their daily activities. This concept, among others, regulates the wearer's willingness to wear a wearable device. Although the comfort-of-wear of current rigid wearable devices (e.g., wristwatches or rings) is partially achieved by their small size, it is not enough for expanding the usage of wearable devices. For example, some people choose not to wear a watch or a wedding ring because of how it feels to wear. Small children or people with dementia tend to remove any irregular things (e.g., healthcare sensors and jewelry) on their body as their importance is difficult to be understood. Besides, the small area of current wearable devices limits the interaction with the wearers (e.g., display size and number of sensors). It is necessary to develop engineering approaches to make comfort-of-wear possible even if the device area is large. We predict that comfort-of-wear will be one of the deciding criteria for the further proliferation of next-generation healthcare monitoring devices (**Figure 1**a).

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REVIEW

Technology Roadmap for Flexible Sensors

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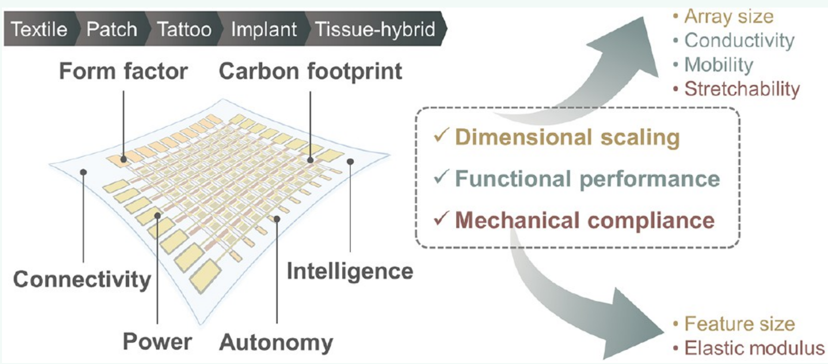

ABSTRACT: Humans rely increasingly on sensors to address grand challenges and to improve quality of life in the era of digitalization and big data. For ubiquitous sensing, flexible sensors are developed to overcome the limitations of conventional rigid counterparts. Despite rapid advancement in bench-side research over the last decade, the market adoption of flexible sensors remains limited. To ease and to expedite their deployment, here, we identify bottlenecks hindering the maturation of flexible sensors and propose promising solutions. We first analyze

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