シンポジウム(口頭講演) | シンポジウム：先端イオン/電子顕微鏡技術のナノスケール材料・デバイスへの応用展開

2024年9月18日(水) 13:30 ~ 18:15 A36 (朱鷺メッセ3F)

[18p-A36-1~11] 先端イオン/電子顕微鏡技術のナノスケール材料・デバイスへの応用展開

小川 真一(産総研)、 大塚 祐二(東レリサーチセンター)

英語発表

13:30 ~ 13:45

[18p-A36-1]

Application of Advanced ion / electron microscopy for future nano scale materials and devices -Introduction to the Symposium-

〇Shinichi Ogawa[1], Jun Taniguchi[2] (1.AIST, 2.TUS)

英語発表

13:45 ~ 14:15

[18p-A36-2]

From Dual Damascene to Semi-Damascene and new materials: opportunities for characterization in interconnects

〇Zsolt Tokei[1] (1.IMEC)

英語発表

14:15 ~ 14:45

[18p-A36-3]

Evaluation of hydrogen-gas-field-ionization ion source and its application

〇Shinichi Matsubara[1], Hiroyasu Shichi[1], Tomihiro Hashizume[2] (1.Hitachi, Ltd. CDS, 2.Hitachi, Ltd. CER)

英語発表

14:45 ~ 15:15

[18p-A36-4]

Nanoscale High-Transition Temperature Josephson Junctions and SQUIDs

〇Shane Cybart[1] (1.UC Riverside)

英語発表

15:15 ~ 15:45

[18p-A36-5]

Nanosized quantum sensor spots in hexagonal boron nitride created using helium ion microscopy

〇Kento Sasaki[1] (1.UTokyo)

英語発表

16:00 ~ 16:15

[18p-A36-6]

Charge trap memory based on MoS_2 with He^+-irradiated h-BN as a trapping layer

〇Mahito Yamamoto[1], Takuya Iwasaki[2], Keiji Ueno[3], Takashi Taniguchi[2], Kenji Watanabe[2], Yukinori Morita[4], Shinichi Ogawa[4], Yutaka Wakayama[2], Shu Nakaharai[5] (1.Kansai Univ., 2.NIMS, 3.Saitama Univ., 4.AIST, 5.Tokyo Univ. Tech.)

英語発表
16:15 ~ 16:45
[18p-A36-7]
Graphene phononic devices for thermal rectification with He Ion beam technology
〇Fayong Liu[1], Kaidi Sun[1], Qianyu Jia[1], Haiyong Zheng[1], Manoharan Muruganathan[2], Hiroshi Mizuta[2] (1.Ocean Univ. of China, 2.JAIST)

英語発表
16:45 ~ 17:00
[18p-A36-8]
Direct Patterning in Ultrathin Silicon Nanosheets Utilizing Helium Ion Beam Irradiation
〇Yukinori Morita[1], Kensuke Inoue[2], Ryuichi Sugie[2], Shinichi Ogawa[1] (1.AIST, 2.TRC)

英語発表
17:00 ~ 17:30
[18p-A36-9]
In-situ and precise atomic-scale transmission electron microscopy for electronic materials
〇Yukio Sato[1] (1.Kumamoto Univ.)

英語発表
17:30 ~ 17:45
[18p-A36-10]
Mapping Dielectric Response of Materials by Time-Resolved Electron Holography
〇Yoh Iwasaki[1], Toshiaki Tanigaki[2], Keiko Shimada[1], Ken Harada[1], Daisuke Shindo[1] (1.RIKEN, 2.Hitachi, Ltd.)

英語発表
17:45 ~ 18:15
[18p-A36-11]
Characterization of monolayer film with an advanced ULV-SEM
〇Takaya Nakamura[1], Masayasu Nagoshi[1], Kaoru Sato[1], Hiroki Ago[2] (1.JFE Techno-Research Corp., 2.Faculty of Engineering Sciences, Kyushu University)

先端イオン/電子顕微鏡技術のナノスケール材料・デバイスへの応用展開 -シンポジウム開催にあたり-

Application of Advanced ion / electron microscopy for future nano scale materials and devices -Introduction to the Symposium-

産総研 [1]，東京理科大 [2]　○小川 真一 [1]，谷口 淳 [2]
AIST [1], TUS. [2], °Shinichi Ogawa[1], Jun Taniguchi[2]
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This symposium aims to broadly discuss research outcomes and technologies related to the control of nanoscale material properties, the creation and evaluation of devices using advanced gas (helium) ion/electron microscopy techniques. The goal is to deepen understanding of these cutting-edge ion/electron microscopy technologies and to promote their further effective utilization in the research and development of materials and devices. In Japan, the research applications of advanced ion microscopy, particularly scanning gas ion microscopy, remain extremely limited. It is essential to discuss and promote the broader adoption of this technology for its significant potential in physics, chemistry, materials science, and device development. This technique can focus the beam to a diameter as small as 0.35 nm, allowing for the precise control and processing of material properties at the nanoscale. It is a critical technology for the fabrication of novel ultra-fine devices and the manipulation of ultra-fine physical phenomena. Beyond its applications in semiconductors, it offers promising possibilities in thin films, surface science, two-dimensional materials, superconductors, phononics, and various other fields. Moreover, atom probe technology is a precursor to the development of gas ion microscope ion sources and has remarkable applications in the atomic-level analysis of impurities in semiconductor microstructures. On the other hand, recent advancements in electron microscopy technology used for evaluating ultra-fine nanostructures have been impressive. This symposium will also discuss atomic-resolution scanning transmission electron microscopy (STEM) and ultra-low acceleration electron microscopy (SEM) technologies.

Related symposia have been continuously held during the Spring and Fall Meetings of 2017, the Fall Meeting of 2018, the Spring Meeting of 2019, and the Spring Meeting of 2020. We aim to foster deep discussions and encourage participation from experts across various fields. We would also like to express our gratitude to the Nano Charged Particle Beam Industry-Academia Collaboration Committee (ナノ荷電粒子ビーム産学連携委員会) of the Japan Society of Applied Physics for their invaluable cooperation in organizing this event.

From Dual Damascene to Semi-Damascene and new materials: opportunities for characterization in interconnects

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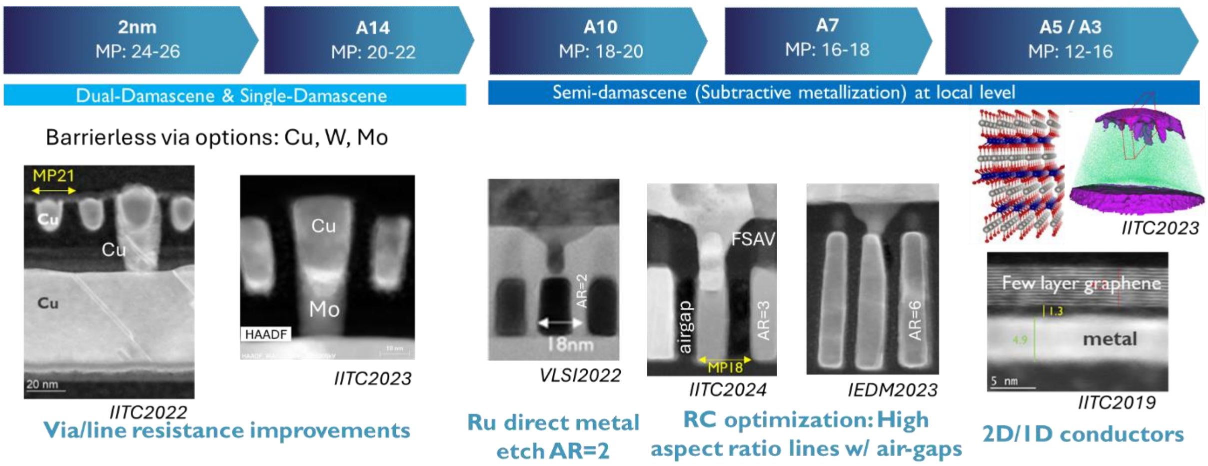
Going forward Back-End-of-Line innovations will be required at design, module and material level. A research roadmap along with pitch scaling at the local level and potential material changes is shown in Figure 1. These options represent a rich variety of tight pitch metal lines and vias -, starting at about 26nm pitch all the way down to 12nm - combined into a multi-level configuration. Today the state-of-the are interconnects are based on damascene technology. Copper dual damascene is a well-known cost-effective industry standard. Control of both vertical and horizontal resistances are crucial along with matched capacitance to keep RC-delay under control, while maintaining robust mechanical properties compatible with packaging technology. Furthermore, with the emergence of backside interconnect options there is a growing need of better understanding thermal material properties as well. These are multiple parameters to optimize simultaneously. To overcome the challenges, several options are being investigated such as for example single damascene, direct metal etch and semi-damascene modules. Besides copper, tungsten, molybdenum, ruthenium emerges as potential part of the puzzle. Going forward binary and ternary alloys might be necessary and even 2D/1D conductors are of research interest. From characterization point of view this opens a lot of opportunities: thin lamella preparation, high resolution imaging, revealing local information at interfaces, understanding and characterizing defects and their impact on resistance etc.

Figure 1 Interconnect module and material options to fuel standard cell scaling; MP: metal pitch



References [1] Zs. Tőkei et al., IEDM2021, [2] K. Croes et al., IEDM 2020, [3,4] M. Van der Veen et al. IITC 2022, IITC2021, [5] G. Murdoch et al., VLSI 2022, [6] A. Gupta et al. IEDM2023, [7] G. Delie et al. IITC2024, [8] S. Achra et al. IITC2020 [9] C. Adelmann et al. IITC 2023, [10] C. Fleischmann et al. IITC2024

水素ガス電界電離イオン源の評価とその応用

Evaluation of hydrogen-gas-field-ionization ion source and its application

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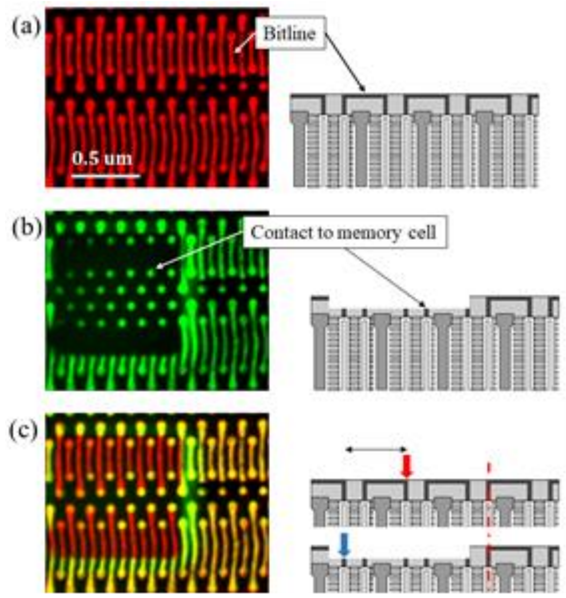
A gas field ionization source scanning ion microscopy (GFIS-SIM) using light element gases, such as helium or hydrogen, is expected to improve the resolution of sample-surface observation because the diffraction aberration is smaller than that of scanning electron microscopy at the same beam acceleration. GFIS-SIM using heavy element gases, such as neon or argon, is also expected to improve processing accuracy compared with conventional focused ion beam (FIB) systems equipped with liquid-metal-ionization sources or plasma-ionization sources.

We present the results of an energy analysis of hydrogen ions by ion beam retarding with an electrostatic lens. Several hydrogen ion species are generated from the hydrogen-ion GIS, among which H_3^+ was shown to be the most suitable for surface observation through this analysis.

We also propose a method of introducing a mixture of light and heavy element gases into the GFIS to eliminate the gas-venting time and speed up the switching of processing and observation functions by changing ion beams. With this method, the beam can be switched from a hydrogen ion beam to neon beam in 0.7 seconds by switching the voltage to the electrode, such as the extraction voltage.

The feature of this switching function is that the two types of beams are emitted from the same column. It is easy to combine the observation and processing functions under optimal conditions for beam focusing (high-resolution conditions). In a multi-column system, e.g., FIB-SEM system, the distance between the objective lens and sample surface (working distance) on the SEM column side must be larger than the optimum value to switch between processing and observation, which forces observation under unfavorable conditions for high-resolution. Our method uses only one identical column for focusing two ion beams, eliminating the above limitation.



(a) Scanning ion microscope (SIM) image before dig process and schematic cross-sectional view of Semiconductor devices with three-dimensional structures (3D NAND) sample. (b) SIM image after dig process (c) Superimposed images

Nanoscale High-Transition Temperature Josephson Junctions and SQUIDs

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Focused helium ion beam nanolithography is a promising approach for fabrication of high-transition temperature superconductor circuits. With this method, a 0.5 nm diameter beam of 40-kV helium ions is scanned over the material, which undergoes a metal-to-insulator transition, due to ion beam induced disorder. (Figure 1.) Regions converted to insulators can be as narrow as 2 nm and serve as Josephson junctions [1] and SQUIDs [2]. Our laboratory has investigated the helium ion beam process and report the electronic transport properties of Josephson junctions fabricated from a range of different ion doses. At doses below 200 ions /nm the junctions exhibit superconductor-normal metal-superconductor (SNS) properties. These SNS junctions have higher resistances and less excess current than masked ion beam junctions. For higher ion beam doses greater than 300 ions/nm, we observe Josephson junctions with superconductor-insulator-superconductor (SIS) properties. In these devices the voltage state resistance increases with decreasing temperature which suggests that the intrinsic shunt is behaving as an insulator with hopping conduction. We correlate these transport properties with Monte Carlo ion implantation simulations which estimate the extent and spatial distribution of the ion beam induced disorder.

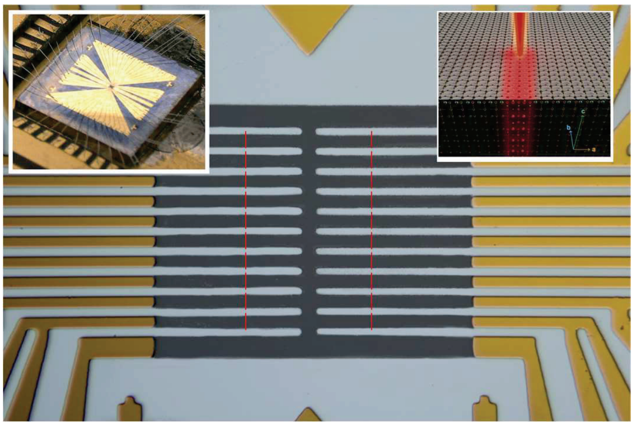

Figure 1. Optical image of a YBCO thin film microbridge chip where the ion beam was scanned across each bridge with different fluence to study how ion dose effects electrical transport. (Left inset) Full view of the bonded 5 mm x 5 mm YBCO on sapphire chip bonded in a chip carrier. (Right Inset) Representation of the helium ion beam interacting with a crystal of YBCO.

[1] S. Cybart, E.Y. Cho, T. Wong, *et al.* "*Nano Josephson superconducting tunnel junctions in* $YBa_2Cu_3O_{7-\delta}$ *directly patterned with a focused helium ion beam*" *Nature Nanotech* **10**, 598–602 (2015). https://doi.org/10.1038/nnano.2015.76
[2] H Li, H Cai, E.Y. Cho *et al* , "*High-transition-temperature nanoscale superconducting quantum interference devices directly written with a focused helium ion beam*", *Appl. Phys. Lett.* 116, 070601 (2020). https://doi.org/10.1063/1.5143026

Nanosized quantum sensor spots in hexagonal boron nitride created using helium ion microscopy

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In this presentation, we introduce a technique for creating quantum sensors using helium ion microscopy, and our recent results [1,2]. Since 2020, a boron vacancy (V_B) defect in hexagonal boron nitride (hBN), a vacancy at a boron site, has been found to function as quantum sensors even at room temperature. By a mechanism similar to the nitrogen vacancy center in a diamond, a representative quantum sensor, the electron spins of this defect can be optically initialized and read out. It allows optical detection of magnetic resonance (ODMR) of the defect's electron spin. In quantum sensing, the magnetic field strength is determined based on the Zeeman effect from the resonance frequencies obtained by ODMR. This quantum sensor works even when the hBN flakes are less than 100 nm thick. It provides nanometer proximity to measurement targets with the van der Waals forces. This proximity is essential to take advantage of the sub-nanometer size of quantum defect sensors to obtain high sensitivity and spatial resolution.

V_B can be created by irradiating hBN crystals with ions, neutrons, or electron beams. Typically, irradiation is uniform and quantum sensors are produced with uniform areal density throughout the flake. In such case, the spatial resolution of the magnetic field imaging is limited to the optical resolution of ODMR system. It is typically about 1 μm, which is more significant than the hBN flake thickness; even if the sensors adhere to the measurement target, the static stray magnetic field from the target is spatially averaged. Thus, the excellent adhesion property of the hBN quantum sensor cannot be fully utilized.

To mitigate this issue, we arrange nanosized sensor spots using helium ion microscopy (HIM) [1,2]. HIM is a microscope that utilizes focused helium ion beams to enable resolutions of up to sub-nanometers. As a proof-of-principle, we used HIM to irradiate He ions to spot sizes of less than $(100\ \text{nm})^2$ to create quantum sensor spots. We demonstrated that magnetic field imaging with higher spatial resolution by arranging sensor spots in an array [1]. We also characterized quantum sensors created with different hBN film thicknesses, substrate surfaces, and ion dosages. The obtained results were discussed to obtain the irradiation conditions that maximize the sensitivity [2]. Our creation technique can be applied to 0-dimensional objects, such as quantum dots, and symmetric or periodic structures, such as magnetic domains and superconducting vortices.

We would like to thank Mr. Tomohiko Iijima, operator, for the use of the helium ion microscope in the Super Clean Room at the National Institute of Advanced Industrial Science and Technology (AIST).

[1] K. Sasaki, Y. Nakamura, H. Gu, M. Tsukamoto, S. Nakaharai, T. Iwasaki, K. Watanabe, T. Taniguchi, S. Ogawa, Y. Morita, and K. Kobayashi, Appl. Phys. Lett. **122**, 244003 (2023).

[2] H. Gu, M. Tsukamoto, Y. Nakamura, S. Nakaharai, T. Iwasaki, K. Watanabe, T. Taniguchi, S. Ogawa, Y. Morita, K. Sasaki, and K. Kobayashi, *to be submitted*.

Charge trap memory based on MoS_2 with He^+-irradiated h-BN as a trapping layer

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Charge trap memory has attracted much attention for applications in memory-based computing. Two-dimensional (2D) materials such as graphene and transition metal dichalcogenides have great potential as building blocks of charge trap memory, owing to the absence of surface dangling bonds and the stacking degree of freedom. However, wide range control of trap density is challenging with an intrinsic 2D material, although is crucial for novel analog computing applications. Here, we fabricated MoS_2-based charge trap memory using He^+-irradiated h-BN as a charge trapping layer, where the trap density can be tuned widely with the He^+ dose amount. BN flakes were exfoliated from bulk crystals onto Si/SiO_2(285 nm) and irradiated with He^+ by using a helium ion microscope. After the irradiation, MoS_2 flakes were transferred onto He^+-irradiated h-BN and Ti/Au electrodes were defined by the electron-beam lithography method (Fig. 1a). The trapping properties of BN-supported MoS_2 were investigated at room temperature in vacuum using the field effect geometry, where Si and SiO_2 serve as a gate and a gate dielectric. Figures 1b-c show transfer characteristics of FETs based on MoS_2 on He^+-irradiated BN with different dose amounts (D). The FETs show hysteresis in the forward and backward gate sweeps, hence working as charge trap memory. With increasing the dose amount, the hysteresis window monotonically widens, indicating that traps are present at defective sites in h-BN. However, at D ~ 10^{16} cm^{-2}, the off state was suppressed, possibly due to the significant increase in the trap capacitance, which may be unfavorable for applications in charge trap memory. Our results could be a guidance to create a charge trapping layer with the controlled trap density for memory applications. ***Acknowledgement*****:** This work was partly supported by the JST SICORP program (grant No. JPMJKB2103) and ARIM of MEXT (JPMXP1224NM5118). We extend our gratitude to Mr. Tomohiko Iijima (AIST) for operating the AIST SCR HIM during the helium ion irradiation process.

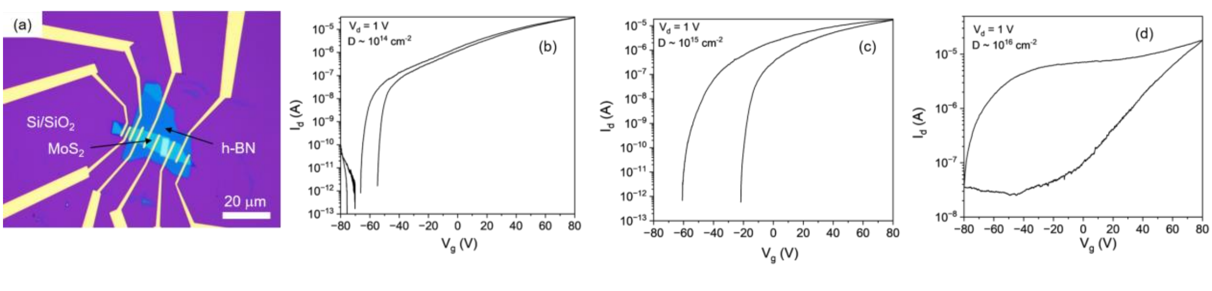

Figure 1. (a) Optical image of MoS_2 on He^+-irradiated h-BN on a Si/SiO_2 substrate. (b)-(d) Drain-current (I_d)-gate voltage (V_g) characteristics of MoS_2 on He^+-irradiated h-BN with dose amounts (D) of 10^{14}, 10^{15}, and 10^{16} cm^{-2}. The drain voltages (V_d) are 1 V.

Graphene phononic devices for thermal rectification with He Ion beam technology

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Graphene is one of the most famous representations of two-dimensional materials with its super physical and chemical properties. With the helium ion beam milling (HIBM) technique, the original graphene structure is artificially modified with a nanometer periodical configuration (Fig.1). It forms a 2D phononic crystal structure for phonon engineering to control the phonon transmission with artificial nanostructures [1]. As the phononic crystals introduce the asymmetry in a suspended graphene ribbon, the thermal rectification phenomenon has been observed and investigated.

In this talk, we will demonstrate the graphene nanostructure fabrication processes with the HIBM technique by coupling it with advanced graphene Nano-Electromechanical System (NEMS) technology. It includes how to reshape the suspended graphene ribbon [2] and pattern periodical nanopores with a 6 nm diameter on a graphene ribbon [3]. With the established experience above, asymmetric graphene phononic crystal structures are introduced on a suspended graphene ribbon. With the help of a differential thermal leakage method, the thermal rectification phenomenon is observed with up to 60% thermal rectification ratio at 150 K [4].

In order to understand the mechanism of thermal rectification on graphene phononic devices, we deeply investigate the phonon transport behavior from each part of the device composition, especially the graphene-gold interface (Fig.2) and asymmetric graphene structures (Fig.3). We use both the molecular dynamics simulation method and the finite element method to investigate the wave properties of the phonon, and found that each part of the device composition gives different weights of the contributions for the thermal rectification phenomenon. This work provides both experimental and theoretical support for further developing graphene-based thermal management devices.

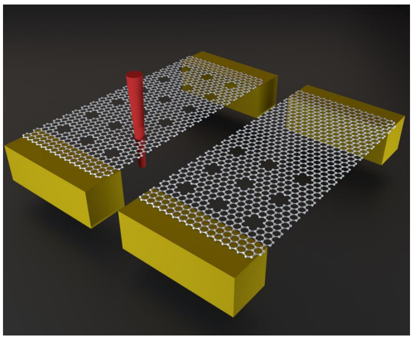

Fig. 1: Schematic illustration of the graphene phononic devices

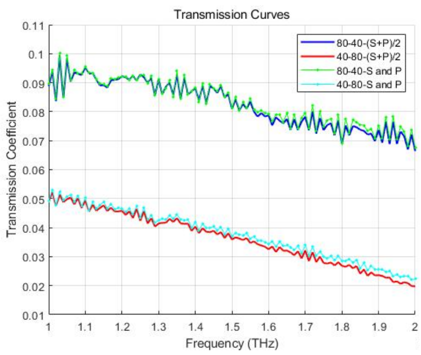

Fig. 3: Wave phonon transmission properties on asymmetric graphene devices

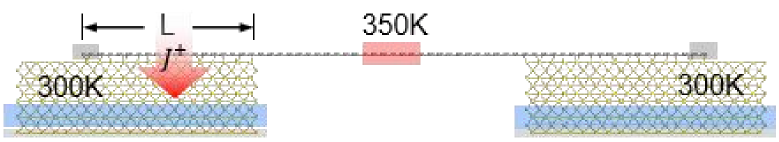

Fig. 2: Molecular dynamics simulation to investigate the interface heat transfer properties.

Acknowledgement:
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Reference:

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2. M. E. Schmidt, H. Mizuta, et al., Small 15, 46, 1903025 (2019)
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4. F. Liu, H. Mizuta, et al., Nano Futures 5 045002 (2021)

Direct Patterning in Ultrathin Silicon Nanosheets Utilizing Helium Ion Beam Irradiation

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Helium Ion Microscopy (HIM) has been used as an electron microscopy using secondary electrons generated by focused helium (He) ion beam irradiation onto the sample. Using ion collision phenomena, it is also possible to modify material properties and perform etching processing by tuning the energy and dose of the He^+ beam. In this work, ultra-thin silicon nanosheets were irradiated with the focused He^+ beam and aim to perform nanofabrication with nanometer-level position control without using lithography technology. Using an extremely uniformly thinned silicon layer, we verified the possibility of forming nanopore arrays with positional control, like two-dimensional materials [1].

A (100) oriented silicon-on-insulator (SOI) sample was used for the experiment. The SOI was etched with oxygen to a thickness of approximately 1 to 3 nm [2]. The He^+ beam was focused at approximately 0.35 nm and the acceleration voltage was 30 keV, and changes in processed shape and defect generation by modulating the dose were investigated. Fabricated shapes were evaluated using a secondary electron image of the sample using HIM.

Figure 1 is a typical HIM image of the sample surface irradiated with 7 × 7 of 20 nm rectangular dots at a pitch of 100 nm. A bright spot is observed at the irradiation position, and as the dose increases, the central part of the irradiation area changes into a “concave” shape. Overall, swelling due to blistering [3] caused by helium gas injection is observed, noticeable over 1e19 cm^{-2} irradiation. The photoluminescence measurement revealed that before and after nanopore formation no significant increase in defects was observed due to ion irradiation. When the SOI layer is made even thinner, the dose required to create the depressions shown in Fig. 1 decreases, suggesting that He^+ beam irradiation realizes nanofabrication of the ultrathin Si layer.

This work was partly supported by JSPS KAKENHI, Grant Number 22K18799.

[1] F. Liu et al., Nano Futures 5 (2021) 045002.
[2] Y. Morita et al., IEDM 2015, 390.
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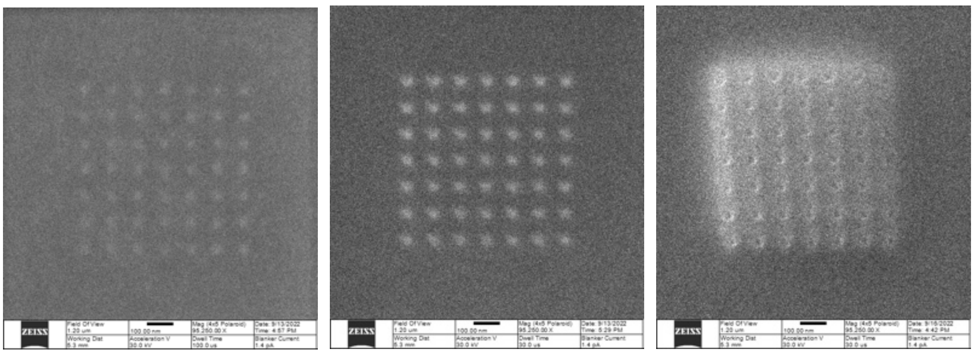

Fig. 1 HIM images after HIM nanolithography on very thin (3.6 nm) SOI. The image area is 1200 × 1200 nm. (a) 1e17, (b) 1e18, (c) 1e19 cm^{-2} of He^+ dose, respectively.

In-situ and precise atomic-scale transmission electron microscopy for electronic materials

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Intriguing physical properties of materials often originate from their micro and nano-scale structures and their responses to external stimuli. In this context, we explore dielectric materials as a prime example. The emergence of dielectricity, piezoelectricity, and ferroelectricity is closely linked to the intricate details of the crystal unit cell and the structure of domains where the polarization of numerous unit cells is aligned in the same direction. Moreover, the response of these structures to applied external electric fields plays a significant role. (Scanning) Transmission electron microscopy is a powerful method that enables us to elucidate detailed unit-cell and domain structures, along with their responses to an electric field, with high spatial resolution and in real time. In this talk, we will introduce some of our key results.

The atomic arrangement of crystals can be directly observed using atomic-scale STEM (scanning transmission electron microscopy). However, errors in the obtained internal atomic positions within unit cells and lattice parameters are usually not negligible. We have developed a methodology that significantly reduces these errors. Errors in lattice parameters can be reduced from 2−3% to 0.1%, and those in cell angles from approximately 0.6° to around 0.1° [1,2]. This method can be applied to determine or discover new crystal phases in localized regions.

In-situ electrical biasing transmission electron microscopy (TEM) has been utilized to visualize response of ferroelectric domain structure by applying electric fields. For example, it was clarified that lamellar-like nanodomains in a piezoelectric single crystal, PMN-PT ($0.68Pb(Mg_{1/3}Nb_{2/3})O_3-0.32PbTiO_3$) reorients by applying an electric field and revert to an original state when the field is removed. These results can explain a part of piezoelectric coefficient and the low hysteresis in the strain-electric-field loop [3]. Not only direct-current electric field but also alternative-current field can be applied using our system. Furthermore, such in-situ observations can be done at an atomic scale precisely by combining the aforementioned methodology [2,4].

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Acknowledgement

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時間分解電子線ホログラフィーによる 画像化誘電分光法の試み

Mapping Dielectric Response of Materials by Time-Resolved Electron Holography

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電気化学素子の解析に多用される誘電分光 または AC インピーダンス測定は，２端子間の試料を流れる電流 vs.電圧の周波数特性を調べる手法である。測定結果を再現する等価回路の考察を経て試料の内部構造への洞察が得られるので，広範な応用がある。いっぽう電子線ホログラフィーは，電子顕微鏡中で試料ならびに周りの空間の電位分布を画像化する手法として知られる。ならば，これを交流電圧下の試料に適用することにより「誘電応答の空間分布をより直接的に観測できないか」と考えた。(1)

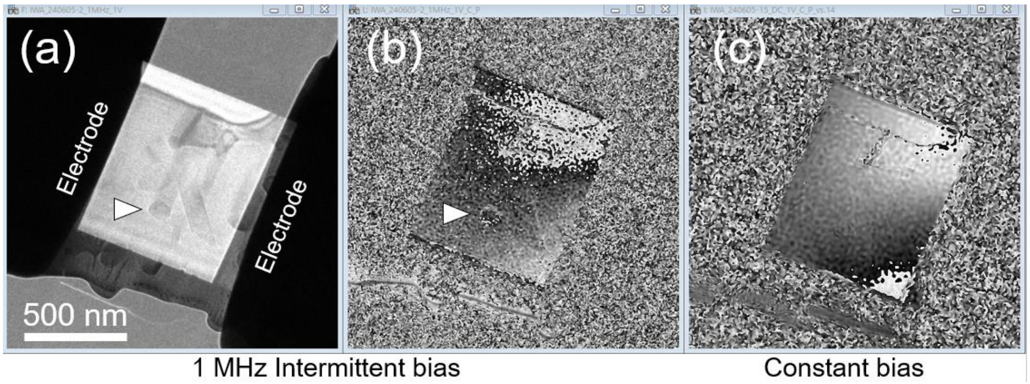

Fig.1 (a) Interference image obtained with accumulated exposures synchronized to the periods with +1V bias voltage on the specimen repeated at 1MHz. (b) Phase image reconstructed from the interference image, (a). (c) Phase image reconstructed from another interference image (not shown) obtained under +1 V constant bias.

Li^+イオン伝導性のガラスセラミック材料を薄片化し，ふたつの金属電極間に保持して観測対象とした。加速電圧 300kV の透過電子顕微鏡（HF-3000X, Hitachi, Ltd.）の電子ビームを ON/OFF してストロボスコピックな露光を行なった。電極間に周波数 1 MHz で +/-1V の矩形波交番電圧を印加し，+1V が印加される期間に同期した露光を積算して得た干渉像を Fig.1(a)に，これに基づいて再生した位相像を Fig.1(b)に示した。また，DC 1V を印加して連続露光で撮影した干渉像に基づく位相像を Fig.1(c)に示した。DC 印加時（c）の位相分布は平坦であるが，同じ 1V でも間欠印加中に撮影した場合の位相分布（b）には矢印▷の先に輪郭を持つ斑点が認められ，1MHz の間欠電場には追随できていない分極成分があることがわかる。この斑点には対応する試料内部の粒構造が干渉像(a)に認められ,この領域の誘電応答が周囲と異なることが示唆される。

ストロボ露光を利用した電子線ホログラフィーによって，試料内部の誘電応答の不均一を可視化する可能性が示された。観測に利用できる周波数域の広さが重要であるが，先に提案した差動ブランキング（Differential Blanking）法(2) を用いてビームを ON/OFF する間欠露光によれば，時間幅が最短 20 ナノ秒までの露光を積算して干渉像，位相像が得られている。 ［ 本研究は科研費の助成（21K04892）を受けて行なわれた。］

(1) Y. Iwasaki, Z. Akase, K. Shimada, K. Harada, and D. Shindo, “Time-Resolved Electron Holography and Its Application to an Ionic Liquid Specimen”, Microscopy, **72** (2023) 455-459.
(2) International Patent Application: PCT/JP2022/011544.

Characterization of monolayer film with an advanced ULV-SEM

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Two-dimensional (2D) materials with atomic-level thicknesses are attracting attention because of their novel electronic, optical, and magnetic properties [1]. It is important to control the number of layers, composition, and structure of these 2D materials, and thus a method to evaluate them quantitively and easily is required. Although scanning electron microscopy (SEM) is widely used for microscopic evaluation of materials, conventional SEM is not suitable for characterizing 2D materials. In recent years, SEM and related techniques have made remarkable progress. For example, ultra-low accelerating voltage SEM (ULV-SEM) enables surface-sensitive observation. A new windowless energy-dispersive X-ray spectrometer (EDX) allows elemental analysis under the same conditions where surface-sensitive observation is performed [2]. We have been working on the evaluation of various advanced materials by making full use of these techniques [3]. In this study, we characterized 2D materials such as graphene oxide (GO) [4] and molybdenum disulfide (MoS_2) using the latest ULV-SEM equipped with a windowless EDX.

Figure 1 shows backscattered electron (BSE) images and EDX elemental mapping images recorded for a MoS_2 monolayer film with accelerating voltages of 5 kV (a) and 1 kV (b). The monolayer sheet of MoS_2, a kind of transition metal dichalcogenide (TMDC), was deposited on a sapphire substrate using a CVD method and then transferred onto a SiO_2/Si substrate. The MoS_2 film is not visible in the BSE image, and no S or Mo distribution could be found in the EDX mapping with the accelerating voltage of 5 kV. This is because the information depth at 5 kV is much deeper than the thickness of the MoS_2 monolayer film (estimated to be about 0.6 nm). The peak energies of S-K (about 2.31 keV) and Mo-L (about 2.29 keV) are close together, making separation of the peaks difficult with EDX. We think this is also a reason why distribution of S and Mo are difficult to be detected. In the BSE image at 1 kV, MoS_2 films exhibit a brighter contrast on the dark SiO_2/Si substrate. The S-L and Mo-M signals are detected corresponding to the MoS_2 film in the EDX mapping. A decrease of Si signal in the MoS_2 area compared to that in the Si substrate is also observed. The new high-sensitive windowless SDD enables detection of ultrasoft X-rays, such as Si-L (about 0.09 keV), S-L (about 0.15 keV), and Mo-M (about 0.19 keV) lines that cannot be detected with a conventional EDX. The shallow penetration depth of primary electrons and the relatively large energy difference between S-L and Mo-M peaks made it possible to visualize the distribution of S and Mo. This ULV-SEM/windowless EDX will provide characterization of various 2D materials with a high throughput.

References: [1] H. Ago., Oyo Buturi, **90**(2021) 617-622. [2] M. Nagoshi, K. Sato, and T. Aoyama, J. Surf. Anal. **24**(2017) 129-135. [3] T. Nakamura, K. Sato, and M. Nagoshi, J. Surf. Anal. **26**(2019) 206-207. [4] https://www.oxinst.jp/casestudy/jfe-tec

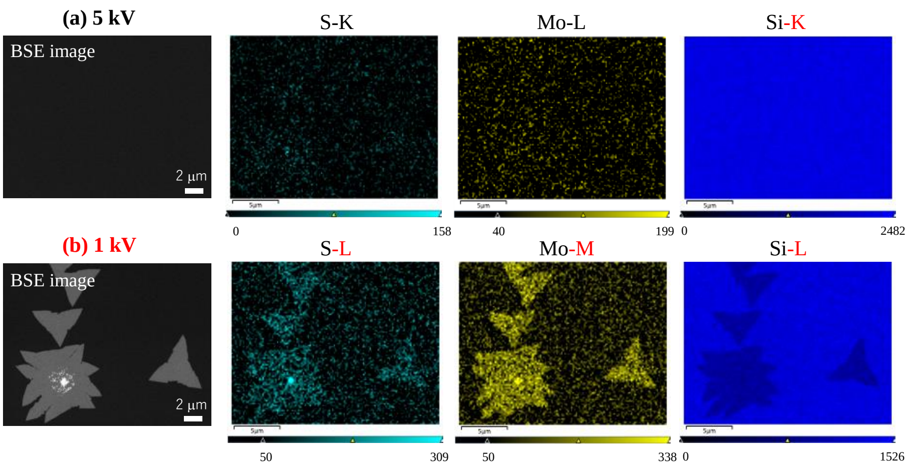

Fig.1 BSE images and EDX mapping results of MoS2 monolayer at (a) 5 kV and (b) 1 kV, respectively.The analytical areas of (a) and (b) are identical.