

***Japan Academy Prize to:***

Yuichi IKUHARA  
Professor, Institute of Engineering  
Innovation, School of  
Engineering, The University of  
Tokyo  
Principal Investigator, Advanced  
Institute for Materials Research,  
Tohoku University  
Visiting Principal Researcher,  
Japan Fine Ceramics Center

and

Naoya SHIBATA  
Director and Professor,  
Institute of Engineering  
Innovation, School of  
Engineering, The University  
of Tokyo



for “Development of State-of-the-Art Electron Microscopy and Contribution to Nano Interface Technology” (Joint Research)

***Outline of the work:***

Dr. Yuichi Ikuhara has been a leading figure in the field of advanced electron microscopy and nanomaterial science in the 21st century. He has made numerous world-leading contributions, and has ascended to an international leadership role in the field. Specifically, he has pioneered the state-of-the-art development of transmission electron microscopy for more than a quarter of a century. Consequently, he has established innovative methods for the quantitative characterization and analysis of local atomic structures, elemental distributions, and local electronic states at nanointerfaces and lattice defects, both experimentally and computationally.

One of his specific research achievements is the direct observation of atoms related to material functions, such as dopants and impurities at the material interface, using ultra-high resolution scanning transmission electron microscopy (STEM). This study has facilitated a major breakthrough in the field of material science. He was the first in Japan to introduce spherical aberration-corrected STEM for analyzing material interfaces. In addition, he proposed a method that combined electron energy loss spectroscopy (EELS) at the atomic-resolution level with first-principles calculations, and established a quantitative analysis method for atomic structures and electronic states, such as interfaces and dislocations.

In 2017, Dr. Ikuhara collaborated with an electron microscope manufacturer to achieve a spatial

resolution of 40.5 pm for STEM, thus setting a world record for the highest performance of a STEM at that time. They also proposed and developed a new annular bright-field (ABF)-STEM technique that facilitated selective capture of electrons transmitted and/or scattered at low angles. This method enabled the first direct observation of lithium atoms in lithium-ion battery materials and hydrogen atom columns in hydrogen storage materials, which have attracted attention as energy and environmental materials. This research led to the worldwide diffusion of this method as a new analytical technique that originated from Japan, which is now routinely used as a general method in overseas research institutes.

Dr. Naoya Shibata has led the world in the advancement of atomic-resolution STEM and its application to the study of material interfaces. Specifically, he has pioneered the development of a method for the direct visualization of electromagnetic field distribution at an atomic scale, which has been extensively used as a new electron microscopy technique in the field of materials and devices. He has reported numerous research results that have contributed to the true and deeper understanding of the mechanisms of interface-originated properties in various materials.

A specific example of Dr. Shibata's research is the development of segmented STEM detector for atomic-resolution imaging. This detector has been used to observe the electric field at the interface of a semiconductor p-n junction and the magnetic field inside a magnetic skyrmion using the differential phase contrast method. Furthermore, he modified this method to precisely measure the slight deflection of the electron beam caused by the existing electric field between the positively charged atomic nucleus and negatively charged electron cloud. Due to his novel discovery the electric field distribution inside an atom can be directly observed in real space for the first time in the world. This is an innovative achievement that is expected to facilitate the use of electron microscopes for real-space observation of intra- and interatomic structures, rather than only observe atomic arrangements.

In 2019, in collaboration with an electron microscope manufacturer, Dr. Shibata successfully developed a new objective lens that realized the atomic-resolution observation while maintaining a sample in magnetic-field-free conditions, which had been a relatively unexplored area of electron microscopy. For the first time, a spatial resolution of less than 1 angstrom was achieved in a magnetic field-free environment, thus rendering the observation of the atomic structure of magnetic materials possible. Furthermore, using this electron microscope, he also realized real-space visualization of the intrinsic magnetic field of an antiferromagnet for the first time in 2022. These achievements are considered groundbreaking breakthroughs in the field of electron microscopy and have received high international acclaim.

Dr. Ikuhara and Dr. Shibata have been conducting research to elucidate various material phenomena that have been previously considered black boxes through the appropriate development of state-of-the-art electron microscopy techniques developed in a complementary and collaborative manner. Their studies have been focused on the correlation between interfacial atomic and electronic structures and functional properties; identification of atomic positions of light elements in inorganic materials and their relationship to physical properties; clarification of dislocation cores and lattice defect structures; understanding material deformation and fracture mechanisms via *in situ* observation; perception of segregation mechanisms at ceramic grain boundaries; studying heterogeneous catalytic interface structures; and the discovery of one-dimensional ordered crystals formed at the grain boundary triple point. Based on the results of such studies, they have designed and created new materials that utilize the functions of interfaces and lattice defects. These results have been published in more than 900 original papers in international journals, and have received extensive appreciation worldwide. Thus, these achievements demonstrate the significant contributions made by them to the fields of advanced electron microscopy and nanomaterial science.

### List of Main Publications

- [1] T. Futazuka, R. Ishikawa, N. Shibata, and Y. Ikuhara, “Grain boundary structural transformation induced by co-segregation of aliovalent dopants”, *Nature Communications*, 13, 5299 (2022).
- [2] J. K. Wei, B. Feng, E. Tochigi, N. Shibata, and Y. Ikuhara, “Direct imaging of the disconnection climb mediated point defects absorption by a grain boundary”, *Nature Communications*, 13, 1455 (2022).
- [3] Y. Kohno, T. Seki, S. D. Findlay, Y. Ikuhara, and N. Shibata, “Real-space visualization of intrinsic magnetic field of an antiferromagnet”, *Nature*, 602, 234–239 (2022).
- [4] J. K. Wei, B. Feng, R. Ishikawa, T. Yokoi, K. Matsunaga, N. Shibata, and Y. Ikuhara, “Direct imaging of atomistic grain boundary migration”, *Nature Materials*, 20, 951–955 (2021).
- [5] K. Inoue, K. Kawahara, M. Saito, M. Kotani, and Y. Ikuhara, “3D arrangement of atomic polyhedra in tilt grain boundaries”, *Acta Materialia*, 202, 266–276 (2021).
- [6] E. Tochigi, B. Miao, A. Nakamura, N. Shibata, and Y. Ikuhara, “Atomic-scale mechanism of rhombohedral twinning in sapphire”, *Acta Materialia*, 216, 117137 (2021).
- [7] S. Sasano, R. Ishikawa, G. Sánchez-Santolino, H. Ohta, N. Shibata, and Y. Ikuhara, “Atomistic origin of Li-ion conductivity reduction at  $(\text{Li}_{3x}\text{La}_{2/3-x})\text{TiO}_3$  grain boundary”, *Nano Letters*, 21, 6282–6288 (2021).
- [8] R. Ishikawa, R. Tanaka, K. Kawahara, N. Shibata, and Y. Ikuhara, “Atomic-resolution topographic imaging of crystal surfaces”, *ACS nano*, 15, 9186–9193 (2021).
- [9] K. Nakayama, R. Ishikawa, S. Kobayashi, N. Shibata, and Y. Ikuhara, “Dislocation and oxygen-release driven delithiation in  $\text{Li}_2\text{MnO}_3$ ”, *Nature Communications*, 11, 4452 (2020).
- [10] J. K. Wei, T. Ogawa, B. Feng, T. Yokoi, R. Ishikawa, A. Kuwabara, K. Matsunaga, N. Shibata, and Y. Ikuhara, “Direct measurement of electronic band structures at oxide grain boundaries”, *Nano Letters*, 20, 2530–2536 (2020).
- [11] D. Q. Yin, C. L. Chen, M. Saito, K. Inoue, and Y. Ikuhara, “Ceramic phases with one-dimensional long-range order”, *Nature Materials*, 18, 19–23 (2019).
- [12] N. Shibata, Y. Kohno, A. Nakamura, S. Morishita, T. Seki, A. Kumamoto, H. Sawada, T. Matsumoto, S. D. Findlay, and Y. Ikuhara, “Atomic resolution electron microscopy in a magnetic field free environment”, *Nature Communications*, 10, 2308 (2019).
- [13] S. Kondo, A. Ishihara, E. Tochigi, N. Shibata, and Y. Ikuhara, “Direct observation of atomic-scale fracture path within ceramic grain boundary core”, *Nature Communications*, 10, 2112 (2019).
- [14] B. Feng, R. Ishikawa, A. Kumamoto, N. Shibata, and Y. Ikuhara, “Atomic scale origin of enhanced ionic conductivity at crystal defects”, *Nano Letters*, 19, 2162–2168 (2019).
- [15] H. P. Li, M. Saito, C. L. Chen, K. Inoue, K. Akagi, and Y. Ikuhara, “Strong metal-metal interaction and bonding nature in metal/oxide interfaces with large mismatches”, *Acta Materialia*, 179, 237–246 (2019).
- [16] P. Gao, S. Z. Yang, R. Ishikawa, N. Li, B. Feng, A. Kumamoto, N. Shibata, P. Yu, and Y. Ikuhara, “Atomic-scale measurement of flexoelectric polarization at  $\text{SrTiO}_3$  dislocations”, *Physical Review Letters*, 120, 267601 (2018).
- [17] S. Kobayashi, A. Kuwabara, C. A. J. Fisher, Y. Ukyo, and Y. Ikuhara, “Microscopic mechanism of biphasic interface relaxation in lithium iron phosphate after delithiation”, *Nature Communications*, 9, 2963 (2018).
- [18] S. Morishita, R. Ishikawa, Y. Kohno, H. Sawada, N. Shibata, and Y. Ikuhara, “Attainment of 40.5 pm spatial resolution using 300 kV scanning transmission electron microscope equipped with fifth-order

- aberration corrector”, *Microscopy*, 67, 46–50 (2018).
- [19] N. Shibata, S. D. Findlay, T. Matsumoto, Y. Kohno, T. Seki, G. Sánchez-Santolino, and Y. Ikuhara, “Direct visualization of local electromagnetic field structures by scanning transmission electron microscopy”, *Accounts of Chemical Research*, 50, 1502–1512 (2017).
- [20] E. Tochigi, Y. Kezuka, A. Nakamura, A. Nakamura, N. Shibata, and Y. Ikuhara, “Direct observation of impurity segregation at dislocation cores in an ionic crystal”, *Nano Letters*, 17, 2908–2912 (2017).
- [21] N. Shibata, T. Seki, G. Sánchez-Santolino, S. D. Findlay, Y. Kohno, T. Matsumoto, R. Ishikawa, and Y. Ikuhara, “Electric field imaging of single atoms”, *Nature Communications*, 8, 15631 (2017).
- [22] P. Gao, H. J. Liu, Y. L. Huang, Y. H. Chu, R. Ishikawa, B. Feng, Y. Jiang, Y. Jiang, N. Shibata, E-G. Wang, and Y. Ikuhara, “Atomic mechanism of polarization-controlled surface reconstruction in ferroelectric thin films”, *Nature Communications*, 7, 11318 (2016).
- [23] B. Feng, T. Yokoi, A. Kumamoto, M. Yoshiya, Y. Ikuhara and N. Shibata, “Atomically ordered solute segregation behaviour in an oxide grain boundary”, *Nature Communications*, 7, 11079 (2016).
- [24] S. Kondo, T. Mitsuma, N. Shibata, and Y. Ikuhara, “Direct observation of individual dislocation interaction processes with grain boundaries”, *Science Advances*, 2, e1501926 (2016).
- [25] S. Kobayashi, C. A. J. Fisher, T. Kato, Y. Ukyo, T. Hirayama, and Y. Ikuhara, “Atomic-scale observations of (010) LiFePO<sub>4</sub> surfaces before and after chemical delithiation”, *Nano Letters*, 16, 5409–5414 (2016).
- [26] H. Yu, Y.-G. So, A. Kuwabara, E. Tochigi, N. Shibata, T. Kudo, H. Zhou, and Y. Ikuhara, “Crystalline grain interior configuration affects lithium migration kinetics in Li-rich layered oxide”, *Nano Letters*, 16, 2907–2915 (2016).
- [27] Z. Wang, M. Saito, K. P. McKenna, S. Fukami, H. Sato, S. Ikeda, H. Ohno, and Y. Ikuhara, “Atomic-scale structure and local chemistry of CoFeB-MgO magnetic tunnel junctions”, *Nano Letters*, 16, 1530–1536 (2016).
- [28] R. Sun, Z. Wang, M. Saito, N. Shibata, and Y. Ikuhara, “Atomistic mechanisms of nonstoichiometry-induced twin boundary structural transformation in titanium dioxide”, *Nature Communications*, 6, 7120 (2015).
- [29] C. L. Chen, Z. C. Wang, F. Lichtenberg, Y. Ikuhara, and J. G. Bednorz, “Patterning oxide nanopillars at the atomic scale by phase transformation”, *Nano Letters*, 15, 6469–6474 (2015).
- [30] C. Chen, Z. Wang, T. Kato, N. Shibata, T. Taniguchi, and Y. Ikuhara, “Misfit accommodation mechanism at the heterointerface between diamond and cubic boron nitride”, *Nature Communications*, 6, 6327 (2015).
- [31] R. Ang, Z. C. Wang, C. L. Chen, J. Tang, N. Liu, Y. Liu, W. J. Lu, Y. P. Sun, T. Mori, and Y. Ikuhara, “Atomistic origin of an ordered superstructure induced superconductivity in layered chalcogenides”, *Nature Communications*, 6, 6091 (2015).
- [32] S.-Y. Choi, S.-D. Kim, M. Choi, H.-S. Lee, J. Ryu, N. Shibata, T. Mizoguchi, E. Tochigi, T. Yamamoto, S.-J. L. Kang, and Y. Ikuhara, “Assessment of strain-generated oxygen vacancies using SrTiO<sub>3</sub> bicrystals”, *Nano Letters*, 15, 4129–4134 (2015).
- [33] L. Gu, D. Xiao, Y.-S. Hu, H. Li, and Y. Ikuhara, “Atomic-scale structure evolution in a quasi-equilibrated electrochemical process of electrode materials for rechargeable batteries”, *Advanced Materials*, 27, 2134–2149 (2015).
- [34] C. Chen, Z. Wang, M. Saito, T. Tohei, Y. Takano, and Y. Ikuhara, “Fluorine in shark teeth: Its direct atomic-resolution imaging and strengthening function”, *Angewandte Chemie-International Edition*, 53, 1543–1547 (2014).
- [35] K. P. McKenna, F. Hofer, D. Gilks, V. K. Lazarov, C. Chen, Z. Wang, and Y. Ikuhara, “Atomic-scale

- structure and properties of highly stable antiphase boundary defects in  $\text{Fe}_3\text{O}_4$ ”, *Nature Communications*, 5, 5740 (2014).
- [36] Z. Wang, M. Saito, K. P. McKenna, and Y. Ikuhara, “Polymorphism of dislocation core structures at the atomic scale”, *Nature Communications*, 5, 3239 (2014).
- [37] R. Ishikawa, N. Shibata, F. Oba, T. Taniguchi, S. D. Findlay, I. Tanaka, and Y. Ikuhara, “Functional complex point-defect structure in a huge-size-mismatch system”, *Physical Review Letters*, 110, 065504 (2013).
- [38] H. Moriwake, A. Kuwabara, C. A. J. Fisher, R. Huang, T. Hitosugi, Y. H. Ikuhara, H. Oki, and Y. Ikuhara, “First-principles calculations of lithium-ion migration at a coherent grain boundary in a cathode material,  $\text{LiCoO}_2$ ”, *Advanced Materials*, 25, 618–622 (2013).
- [39] A. Nakamura, T. Mizoguchi, K. Matsunaga, T. Yamamoto, N. Shibata, and Y. Ikuhara, “Periodic nanowire array at the crystal interface”, *ACS Nano*, 7, 6297–6302 (2013).
- [40] I. Sugiyama, N. Shibata, Z. Wang, S. Kobayashi, T. Yamamoto, and Y. Ikuhara, “Ferromagnetic dislocations in antiferromagnetic  $\text{NiO}$ ”, *Nature Nanotechnology*, 8, 266–270 (2013).
- [41] S.-Y. Chung, S.-Y. Choi, S. Lee, and Y. Ikuhara, “Distinct configurations of antisite defects in ordered metal phosphates: Comparison between  $\text{LiMnPO}_4$  and  $\text{LiFePO}_4$ ”, *Physical Review Letters*, 108, 195501 (2012).
- [42] N. Shibata, S. D. Findlay, Y. Kohno, H. Sawada, Y. Kondo, and Y. Ikuhara, “Differential phase-contrast microscopy at atomic resolution”, *Nature Physics*, 8, 611–615 (2012).
- [43] Y. Ikuhara, “Grain boundary atomic structures and light-element visualization in ceramics: combination of Cs-corrected scanning transmission electron microscopy and first-principles calculations”, *Journal of Electron Microscopy*, Review paper, 60(Supplement 1), S173–S188 (2011).
- [44] Z. Wang, M. Saito, K. P. McKenna, L. Gu, S. Tsukimoto, A. L. Shluger, and Y. Ikuhara, “Atom-resolved imaging of ordered defect superstructures at individual grain boundaries”, *Nature*, 479, 380–383 (2011).
- [45] S. D. Findlay, T. Saito, N. Shibata, Y. Sato, J. Matsuda, K. Asano, E. Akiba, T. Hirayama, and Y. Ikuhara, “Direct imaging of hydrogen within a crystalline environment”, *Applied Physics Express*, 3, 116603 (2010).
- [46] Z. Wang, M. Okude, M. Saito, S. Tsukimoto, A. Ohtomo, M. Tsukada, M. Kawasaki, and Y. Ikuhara, “Dimensionality-driven insulator-metal transition in A-site excess non-stoichiometric perovskites”, *Nature Communications*, 1, 106 (2010).
- [47] N. Shibata, Y. Kohno, S. D. Findlay, H. Sawada, Y. Kondo, and Y. Ikuhara, “New area detector for atomic-resolution scanning transmission electron microscopy”, *Journal of Electron Microscopy*, 59, 473–479 (2010).
- [48] S.-Y. Choi, S.-Y. Chung, T. Yamamoto, and Y. Ikuhara, “Direct determination of dopant site selectivity in ordered perovskite  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  polycrystals by aberration-corrected STEM”, *Advanced Materials*, 21, 885–889 (2009).
- [49] N. Shibata, S. D. Findlay, S. Azuma, T. Mizoguchi, T. Yamamoto, and Y. Ikuhara, “Atomic-scale imaging of individual dopant atoms in a buried interface”, *Nature Materials*, 8, 654–658 (2009).
- [50] N. Shibata, A. Goto, K. Matsunaga, T. Mizoguchi, S. D. Findlay, T. Yamamoto, and Y. Ikuhara, “Interface structures of gold nanoparticles on  $\text{TiO}_2$  (110)”, *Physical Review Letters*, 102, 136105 (2009).
- [51] Y. Ikuhara, “Nanowire design by dislocation technology”, *Progress in Materials Science*, 54, 770–791 (2009).
- [52] S.-Y. Chung, S.-Y. Choi, T. Yamamoto, and Y. Ikuhara, “Atomic-scale visualization of antisite defects

- in  $\text{LiFePO}_4$ ”, *Physical Review Letters*, 100, 125502 (2008).
- [53] N. Shibata, A. Goto, S. Y. Choi, T. Mizoguchi, S. D. Findlay, T. Yamamoto, and Y. Ikuhara, “Direct imaging of reconstructed atoms on  $\text{TiO}_2$  (110) surfaces”, *Science*, 322, 570–573 (2008).
- [54] N. Shibata, M. F. Chisholm, A. Nakamura, S. J. Pennycook, T. Yamamoto, and Y. Ikuhara, “Nonstoichiometric dislocation cores in alpha-alumina”, *Science*, 316, 82–85 (2007).
- [55] J. P. Buban, K. Matsunaga, J. Chen, N. Shibata, W. Y. Ching, T. Yamamoto, and Y. Ikuhara, “Grain boundary strengthening in alumina by rare earth impurities”, *Science*, 311, 212–215 (2006).
- [56] Y. Sato, J. P. Buban, T. Mizoguchi, N. Shibata, M. Yodogawa, T. Yamamoto, and Y. Ikuhara, “Role of Pr segregation in acceptor-state formation at ZnO grain boundaries”, *Physical Review Letters*, 97, 106802 (2006).
- [57] Y. Ikuhara, H. Nishimura, A. Nakamura, K. Matsunaga, and T. Yamamoto, “Dislocation structures of low-angle and near-Sigma 3 grain boundaries in alumina bicrystals”, *Journal of the American Ceramic Society*, 86, 595–602 (2003).
- [58] A. Nakamura, K. Matsunaga, J. Tohma, T. Yamamoto, and Y. Ikuhara, “Conducting nanowires in insulating ceramics”, *Nature Materials*, 2, 453–456 (2003).