Automated characterization of magnetic materials

Kanta Ono
(Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK))

Future research and development in magnetic materials requires to focus on the following four essential research areas:

1. Automation of research and development or automated scientific discovery
2. Autonomous robotic technology for research and development
3. AI or machine learning technology for magnetic materials research
4. Data collection, integration, and infrastructure for public access

In this presentation, we will discuss these priority research topics, mainly from magnetic materials' characterization viewpoints. A high-throughput material characterization system with quantum beams, such as X-rays and neutrons, leads to a drastic increase in measurements' speed and efficiency. However, we believe that the essence of material characterization is to extract useful information and knowledge for researchers and to automate the research and development process. Only performing high-throughput measurements and collecting a large amount of measurement data and compiling them into a database is not enough to improve materials research efficiency. We will discuss a methodology to maximize the information obtained per time and cost in the measurement [1-4]. While high-throughput measurements are becoming more common, most of the measurement data analysis is done manually by skilled experts, which is a bottleneck in the efficiency of research and development. In addition to freeing researchers from simple tasks to devote themselves to research activities, the measurement and analysis of data will be commoditized so that anyone can perform the measurement and analysis tasks that were previously performed only by skilled experts.

Reference

1) T. Ueno, H. Hino, A. Hashimoto, Y. Takeichi, M. Sawada, and K. Ono, "Adaptive design of an X-ray magnetic circular dichroism spectroscopy experiment with Gaussian process modelling", npj Computational Materials 4, 4 (2018).
2) K. Saito, M. Yano, H. Hino, T. Shoji, A. Asahara, H. Morita, C. Mitsumata, J. Kohlbrecher and K. Ono, "Accelerating small-angle scattering experiments on anisotropic samples using kernel density estimation", Sci. Rep. 9, 1526 (2019)
3) Y. Suzuki, H. Hino, M. Kotsugi and K. Ono, "Automated estimation of materials parameter from X-ray absorption and electron energy-loss spectra with similarity measures" npj Computational Materials 5, 39 (2019).
4) Y. Ozaki, Y. Suzuki, T. Hawai, K. Saito, M. Onishi and K. Ono, "Automated crystal structure analysis based on blackbox optimisation", npj Computational Materials 6, 75 (2020).

Adaptive design of experiments for X-ray magnetic circular dichroism spectroscopy

Tetsuro Ueno
(Quantum Beam Science Research Directorate, National Institutes for Quantum and Radiological Science and Technology (QST))

X-ray microscopy such as transmission X-ray microscopy (TXM) or scanning transmission X-ray microscopy (STXM) is a modern experimental technique to observe magnetic domains with several ten-nm spatial resolutions. Observation of magnetic domains with these techniques is based on X-ray magnetic circular dichroism (XMCD), a phenomenon that the the absorption coefficient at absorption edges of ferromagnetic materials differs for right- or left-handed circularly polarized X-rays.

We demonstrated the quantitative analysis of magnetic domains with XMCD-STXM [1]. STXM experiment was performed at the Photon Factory, High Energy Accelerator Research Organization [2]. STXM images around Sm $M_{4,5}$ absorption edges of $SmCo_5$ permanent magnet for right- and left-handed circularly polarized X-rays, respectively. X-ray absorption and XMCD spectra were obtained for an area of 2.7×1.4 μm^2 by 100 nm steps. Spatial distributions of spin and orbital magnetic moments were obtained by applying magneto-optical sum rules to pixel-by-pixel XAS and XMCD spectra.

Although XMCD-STXM is a powerful tool to analyze magnetic domains, the experimental throughput is a problem. Typically, it takes several hours to measure one data set. Experimental parameters for STXM are the number of spatial points (scanning area and steps on sample), the number of energy points (energy range and steps), and the dwell time at each point. To improve the efficiency of STXM, we conceived a reduction of energy points using a machine learning technique, Gaussian process regression. We developed an adaptive design of experiments (ADoE) that combines measurement, analysis and machine learning. It is demonstrated that the ADoE for $Sm_{4,5}$ XMCD spectra reduces the energy points to 20% of a conventional experimental design to obtain magnetic moments with satisfactory accuracy [3].

Reference

1) T. Ueno, A. Hashimoto, Y. Takeichi, and K. Ono, “Quantitative magnetic-moment mapping of a permanent-magnet material by X-ray magnetic circular dichroism nano-spectroscopy”, AIP Advances **7**, 056804 (2017).
2) Y. Takeichi, N. Inami, H. Suga, C. Miyamoto, T. Ueno, K. Mase, Y. Takahashi, and K. Ono, “Design and performance of a compact scanning transmission X-ray microscope at the Photon Factory”, Rev. Sci. Instrum. **87**, 013704 (2016).
3) T. Ueno, H. Hino, A. Hashimoto, Y. Takeichi, M. Sawada, and K. Ono, “Adaptive design of an X-ray magnetic circular dichroism spectroscopy experiment with Gaussian process modelling”, npj Computational Materials **4**, 4 (2018).

Coercivity Analysis based on extended Landau free energy landscape

Masato Kotsugi

(Tokyo University of Science)

Microstructure is an important information that characterizes macroscopic function. The coercivity is a typical issue, and we have analyzed the magnetic domain structure and metallographic structure to discuss the origin of macroscopic coercivity. However, a rather problematic approach has been taken for a long time, in which the results of pinpoint local structural analysis are used to discuss the macroscopic function of the entire system. In other words, most of the information of image data has been discarded and the interpretation of the image data required expert knowledges.

Here, we propose a new energy model that can explain macro functions using entire information of microstructure. Spatial inhomogeneity, which could not be dealt with by the Landau model, is quantified and used as a feature using modern mathematical science. Feature extraction combines Persistent homology, Fourier transformation and Ising model to extract significant Physical Feature in multiscale. Selected features are used to draw the free energy landscape that can explain the magnetization reversal process, then analyze the behavior of the saddle point to discuss the origin of macroscopic coercivity. We design the extended Landau energy model that can handle spatial inhomogeneity and explain the macroscopic functions. The model can connect microscopic microstructure and macroscopic materials' function. Furthermore, the modeling the free energy landscape behind the material functions would allow for analysis that goes into the interpretation of the mechanisms. In this talk, we will introduce our recent research projects related to

(1) Feature extraction from magnetic domain structure using Persistent Homology.
(2) Drawing Extended Landau Free Energy Landscape for the analysis of magnetization reversal process and coercivity.

Reference

T. Yamada, M. Kotsugi et al. Vacuum and Surface Science 62, 153, (2019)
https://doi.org/10.1380/vss.62.153

Drawing the extended Landau free energy landscape of polycrystalline magnetic thin films

Alexandre Lira Foggiatto[1*], Sotaro Kunii[1], Chiharu Mitsumata[2] and Masato Kotsugi[1]
([1]Tokyo University of Science, [2]NIMS)

The understanding of the function of real materials in a heterogeneous system, such as magnetic domain and metallographic structure, has been an outstanding issue in materials science. Thus the development of a consistent and fast analysis method that considers the defects, roughness, crystal sizes, etc. is of utmost importance.1) Here, we are developing a machine learning-based formula that can treat the microscopic morphology and describes the macroscopic properties based on the energy of the system. One important application is to describe the coercivity based on the structure and micromagnetic properties.2) The Landau free energy theory is arduous to be implemented in complex applications due to the pinning de-pinning process of the domain walls.3) Thus, the description of the physics in inhomogeneous polycrystalline systems considering the metallography structure is necessary for advanced material applications.

In this work, we use micromagnetic simulation to calculate the external field dependence of magnetization in polycrystalline permalloy (Fig. 1) and analyze it using unsupervised machine learning to find correlations between the images in the data set. The energy landscape in the magnetization reversal process is successfully visualized as a function of features (Fig. 2). It is an observed correlation between the reduced feature space and the hysteresis loop. The map of the data in lower dimension space of the magnetization, in the same direction of the external magnetic field, displays a clear coercivity dependence. Small grains sizes have smaller components and broader distribution in the feature space, which is inverse proportion to the coercivity. Moreover, the landscape map allows us to access and predict the total energy of the system. Our result implies that the magnetic microstructure can display information about the macroscale properties.

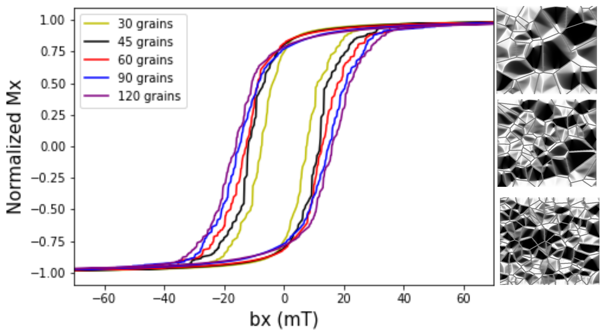

Fig. 1. Hysteresis loop for different grain sizes and magnetic domain structures near coercivity for 30, 60 and 120 grains.

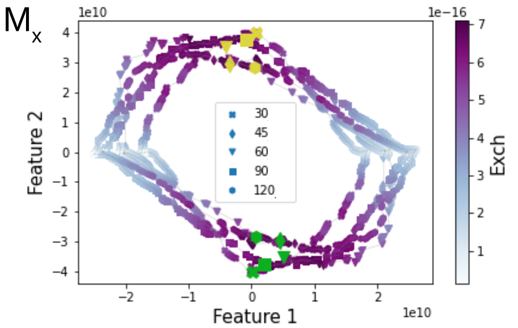

Fig. 2. Reduced feature space of the magnetization reversal process of the x components. The green an yellow points correspond to the positive and negative coercivity.

Reference

1) C. Shen, et al., Acta Materialia **179**, 201 (2019).
2) C. H. Chen, et al., J. Appl. Phys. **93**, 7966 (2003)
3) A. Hubert, R. Schäfer "Magnetic Domains: The Analysis of Magnetic Microstructures" (Springer-Verlag, Berlin, Heidelberg, 1998).

Precision improvement in electron holography: application of information science to magnetic structure analysis

Y. Murakami[1], T. Tanigaki[2], H. Shinada[2] and Y. Midoh[3]
1 Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University,
Fukuoka 819-0395, Japan.
2 Research & Development Group, Hitachi, Ltd., Hatoyama 350-0395, Japan.
3 Graduate School of Information Science and Technology, Osaka University, Suita 565-0871, Japan.

Electron holography, which is a method related to transmission electron microscopy (TEM), can be a tool for the magnetic domain structure analysis, as it enables magnetic flux mapping in a nanometer-scale resolution. Actually, some of the authors 1) have attained the atomic-scale resolution in the flux mapping from an oxide crystal, as it will be briefly mentioned in this symposium. For the applications to materials science and engineering, the other essential factor is "precision" of the magnetic flux density measurement. Importantly, the precision of electron holography depends on the image quality of "hologram" which is made of interference fringes of the incident electrons. (The hologram provides information about the phase shift of the incident electrons which traverse a magnetic specimen.) Although a long-time electron exposure can be an effective way to improve the image quality of holograms, it induces undesired specimen drift during data collection, surface contaminations, radiation damage, and other such problems. We have employed several techniques of information science and/or data science to improve the image quality of holograms. An essential technique is of noise reduction from holograms which were collected in a short exposure time (to suppress the undesired events caused by a long-time exposure). Midoh *et al.* 2) introduced Markov property into the process of noise reduction using the wavelet transform and thresholding. Based on this modeling, they established a criterion for the separation of noise from weak signal in the holograms. The noise reduction improved the precision in phase analysis by 4-5 times as compared with the value from the original (unprocessed) hologram. In addition to this modeling, for another route of the noise reduction, we employed machine learning and the other methods of image processing to carry out the averaging of many holography observations.

Electron holography was applied to the magnetic flux density measurement from a narrow grain boundary produced in a 0.1% Ga-doped Nd-Fe-B sintered magnet 3). Because of the methods of precision improvement, the uncertainty in phase detection was reduced to $2\pi/210$ rad. The result is better than the value ($2\pi/80$ rad) attained in the previous electron holography study which revealed the presence of ferromagnetic grain boundaries in a commercial Nd-Fe-B magnet subjected to the optimal heat treatment 4). A sophisticated electron holography study 3) allowed the magnetic flux density measurements as a function of positions along the grain boundary region: see Fig. 1. The observations provide useful information about the magnetic and/or chemical inhomogeneity in the grain boundary region in the 0.1% Ga-doped Nd-Fe-B magnet.

The authors are grateful to Drs. K. Hono, T. Ohkubo, and T. Sasaki (NIMS) for their collaborations with the study of the 0.1% Ga-doped Nd-Fe-B magnet. This study was supported by CREST (JPMJCR1664) and ESICMM (JPMXP0112101004). A part of the study was supported by FIRST Program initiated by CSTI.

Reference
1) T. Tanigaki *et al.,* manuscript in preparation.
2) Y. Midoh *et al.,* Microsc. **69** (2020) 121.
3) Y. Cho *et al.,* Scripta Mater. **178** (2020) 533.
4) Y. Murakami *et al.,* Acta Mater. **71** (2014) 370.

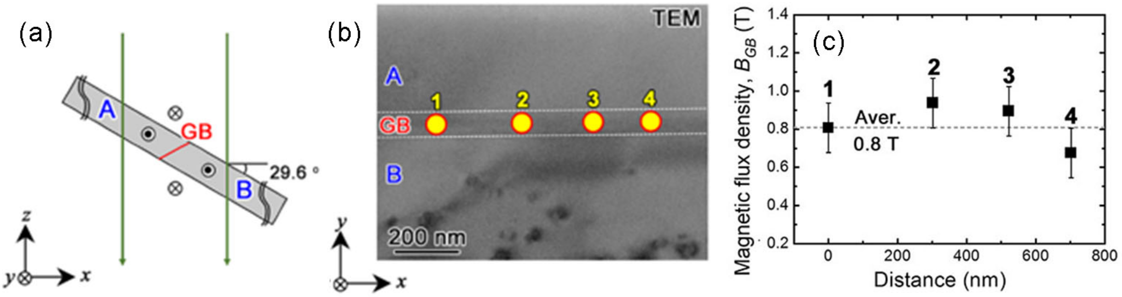

Fig. 1 Magnetic flux density measurements from the grain boundary (GB) in 0.1% Ga-doped Nd-Fe-B sintered magnet. (a) Schematic representation of the cross-section of thin-foil specimen, made of two $Nd_2Fe_{14}B$ grains A and B. (b) TEM image of the thin-foil specimen. Since the plane of GB was tilted away from the incident electron, the projection provides a wide GB region (~90 nm): refer to the area indicated by the white lines. (c) Magnetic flux density measurements from the points 1-4 in (b).