

ISSP - J-PARC Joint Workshop
~ The 16th Korea-Japan meeting on Neutron Science ~
Science Frontier by Neutron Scattering
2018 Jan. 8 - 10

Program and Abstracts

Organized by



The University of Tokyo
The Institute for
Solid State Physics

&



Sponsorship



CROSS



Time table

| | Jan. 8 (Mon) | Jan. 9 (Tue) | Jan. 10 (Wed) |
|---------|--|--|--|
| | Kashiwa Campus Station Satellite | ISSP, UTokyo | ISSP, UTokyo |
| AM | | Registration Opening Session Session I Facility Report Poster Session II Magnetism and Strongly correlated electrons | Session V Instruments, Software and Advanced Data Analysis/ Magnetism and strongly correlated electrons Session VI Magnetism and strongly correlated electrons |
| Noon | | Lunch Business Meeting | Lunch |
| PM | | Poster Session Session III Magnetism and Strongly correlated electrons Session IV Industrial use | Session VII Young scientists/Bio, soft matters and liquid Session VIII Bio, soft matters and liquid Closing Session |
| Evening | Registration Evening session Reception | Banquet | |

Program

Jan. 8th

**Multipurpose space, 2nd Floor, The University of Tokyo Kashiwa Campus Station
Satellite, 178-4, Wakashiba, Kashiwa, Chiba**

Registration

17:00- Registration

Evening session and reception

17:30-19:00

Jan. 9th

**Lecture room, 6th Floor, Institute for Solid State Physics, the University of Tokyo,
Kashiwa, 5-1-5, Kashiwanoha, Kashiwa, Chiba**

Registration

8:30- Registration

Opening

9:00-9:05 Welcome Address by Director

9:05-9:10 Welcome address from Korean side (Sung-Min Choi)

9:10-9:15 Welcome Address by President of JSNS

9:15-9:20 Report on recent J-K collaboration

9:20-10:30 (15min. x 2 + 20min. x 2) Facility reports

Mitsuhiro Shibayama, ISSP. “Current Status of ISSP NSL” (15 min.)

Masayasu Takeda, JAEA, “Current status of JRR3” (15 min.)

Toshiji Kanaya, J-PARC/KEK, “Current status of J-PARC/MLF” (20 min.)

Sungil Park, KAERI, “Current status of HANARO” (20 min.)

10:30-10:45 Coffee break/Poster session I

10:45-12:25(25min. x 4) Magnetism and strongly correlated electrons

Je-Geun Park, Seoul National University, “Magnon-phonon coupling of hexagonal manganites probed by inelastic neutron and X-ray scattering”

Sungdae Ji, Max Planck POSTECH Center, “Kitaev quantum spin liquid and its beyond”

Taro Nakajima, RIKEN, “Small angle neutron scattering study on magnetic skyrmions under various external stimuli”

Ryoichi Kajimoto, J-PARC, “Researches of magnetism and strongly correlated electron systems using the time-of-flight spectrometer 4SEASONS”

12:25-13:00 Lunch Break

(12:25-13:25) Business meeting @ 2nd meeting room

13:00-14:45 Poster session and Coffee

14:45-16:25 (25min. x 4) Magnetism and strongly correlated electrons

Jonathan Leiner, Seoul National University, “Magnetic Excitations of the Cu²⁺ Quantum Spin Chain in Sr₃CuPtO₆”

Haruhiro Hiraka, KAERI, “High-energy magnetic excitation of itinerant-electron antiferromagnet Cr studied on HRC and SIKI”

Minoru Soda, RIKEN, “Magnetic Anisotropy and Magnetoelectric Effect with Spin-Nematic Interaction”

Ki-Yeon Kim, KAERI, “Self-Assembly of Two-dimensional Layered Perovskite Thin Films”

16:25-16:40 Coffee break

16:40-18:20 (25min. x 4) Industrial use

Hyungsub Kim, KAERI, “Thermal phase stability studies on layered cathode materials for Li-ion batteries using combined in-situ high temperature neutron diffraction and gas Analyses”

Youngung Jeong, Changwon University, “Multiaxial flow stress measurement based on X-ray diffraction and uncertainty estimation by incorporating crystal plasticity to Monte Carlo method”

Soo Yeol Lee, Chungnam National University, “Microstructure and Mechanical Properties of AdditiveManufactured Stainless Steel”

Toshirou Tomida, Ibaraki Prefecture, “Industrial Use of Pulsed Neutron Beam Lines Established by Ibaraki Prefecture in J-PARC MLF”

19:00-21:00 Banquet @ lobby

Jan. 10th

Lecture room, 6th Floor, Institute for Solid State Physics, the University of Tokyo,
Kashiwa, 5-1-5, Kashiwanoha, Kashiwa, Chiba

9:00-9:50 (25min. x 2) Instruments, Software and Advanced Data Analysis

Kenji Ohyama, Ibaraki University, “Novel usage of neutrons White neutron holography for investigations of local atomic structures”

Kosuke Hiroi, J-PARC, “Development of a magnetic imaging technique using pulsed polarized neutrons at J-PARC MLF”

9:50-10:40 (25min. x 2) Magnetism and strongly correlated electrons

Shinichi Itoh, KEK, “A trial of observation of quantum phase on HRC”

Sungkyun Park, Pusan National University, “Strain-induced uniaxial magnetic anisotropy variation in epitaxial CoFe₂O₄ thin films”

10:40-11:00 Coffee Break/Photo session

11:00-11:25 (25min. x 1) Magnetism and strongly correlated electrons

June Hyuk Lee, KAERI, “Atomically Engineered Oxide Layers for Novel Magnetoelectric Phenomena”

11:25-12:05 (20min. x 2) Young scientists

Keisuke Matsuura, University of Tokyo, “Spin-Orbital Correlated Dynamics in the Spinel-Type Vanadium Oxide MnV₂O₄”

Shohei Hayashida, ISSP, “Pressure-induced Quantum Phase Transition in Singlet Ground State Magnet CsFeCl₃ Investigated by Neutron Scattering”

12:05-13:05 Lunch break

13:05-14:05 (20min. x 3) Young scientists

Jongsoon Kim, Sejong University, “Development of new 4V-class and zero-strain cathode material for Na ion batteries using neutron diffraction”

SeungHwan Do, Chung-Ang University, “Majorana fermions in the Kitaev quantum spin system α -RuCl₃”

Johannes Reim, Tohoku University, “Spin wave excitations under strong frustration in the layered kagome compound CaBaCo₂Fe₂O₇”

14:05-15:20 (25min. x 3.) Bio, Soft matter and liquid

Soohyung Choi, Hongik University, "Structure of Complex Coacervate Core Micelles"

Koichiro Hori, KEK, "Structure and Mechanical Properties of Elastomer at Surface-modified Carbon Interface"

Tae-Hwan Kim, Chonbuk National University, "Small Angle Scattering Study on Phase Transition of Amphiphilic Molecules in Aqueous Solution"

15:20-15:35 Coffee break

15:35-17:15 (25min. x 4) Bio, Soft matter and liquid

Xiang Li, ISSP, "Structure analysis of critical gelation cluster on the basis of 3D phase diagram"

Takeshi Yamada, CROSS, "Dynamical Behavior of Hydration Water Molecules between Phospholipid Membranes"

Myung Chul Choi, KAIST, "Assembly Architectures of Protein Nanotubes Triggered by Cationic Molecules"

Jaseung Koo, KAERI, "Neutron reflectivity measurements for diffusion dynamics of polymer melts confined between graphene oxide sheets"

17:15-17:30 Closing

Summary of the conference

Announce for next conference and closing remarks

ABSTRACTS

Oral Presentations

Current Status of ISSP-NSL

Mitsuhiro Shibayama

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Since 1961, the ISSP has been playing a central role in neutron scattering activities in Japan not only by performing its own research programs but also by providing a general user program for the university owned various neutron scattering spectrometers installed at the research reactor of JAEA (Tokai). In the JRR-3 reactor (20MW), the university group owns 14 spectrometers, and the Neutron Science Laboratory (NSL) is conducting the general user program. Furthermore the NSL owns state-of-art inelastic neutron scattering spectrometer HRC in J-PARC which started its operation in 2009. On the other hand, ISSP-NSL has been running US-Japan Collaboration Program for Neutron Scattering since 1980s.

However, the Great Earthquake (March 11, 2011) hit East Japan has changed the activities of NSL-ISSP. Due to long shutdown of JRR-3, General User Program has paused since 2011. In order to resume JRR-3, we have various activities, including operation test, improvement and upgrade. On the other hand, NSL-ISSP is running a program to support travel expenses for users who carry neutron scattering experiments at foreign facilities.

The current status and activities of Neutron Science Laboratory, Institute for Solid State Physics, will be overviewed.

Current status of JRR-3

M. Takeda

Materials Sciences Research Center, JAEA, Ibaraki 319-1195, Japan

The large earthquake hit JRR-3 on Mar 11, 2011. JRR-3 itself had no serious damage, but we had to repair the utilities which is necessary for safety operation. We finished the repair and confirmation of the safe operation of JRR-3 within two years after the earthquake.

But because of the tragic nuclear accident of Fukushima Dai-ichi nuclear power plant, the new regulatory requirement was established even for research reactors including JRR-3. We submitted an application document for restarting almost three years ago. But, it has been passed almost 6 years since the earthquake. We are still waiting for the license.

The reviewing process now at last moved to the final stage. JAEA officially announced a schedule for restarting of JRR-3 in June last year. In this schedule, the reactor was expected to restart in February, 2018. Unfortunately, the schedule has to be reconsidered based on the delay of reviewing process. The revised schedule will be announced soon.

Current Status of J-PARC MLF

T. Kanaya

J-PARC MLF

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Japan Proton Accelerator Research Complex (J-PARC) is co-organized by High Energy Accelerator Research Organization (KEK) and Japan Atomic Energy Agency (JAEA) and the accelerator complex consists of 400MeV Linac, 3 GeV rapid cycle synchrotron ring (RCS) and 50 GeV main ring (MR). The accelerated proton beam is injected from the RCS to the Materials and Life Science Experimental Facility (MLF) to produce neutron and muon. The MLF is investigating a wide variety of science such as hard matter, soft matter including, life science, energy materials including batteries and hydrogen adsorbing materials, engineering materials, and high pressure science using neutron and muon.

The MLF was now running at 150 kW until the end of June, 2017 due to the target trouble in 2015 with high operation efficiency more than 90 %. In the summer shutdown, 2017, the target was replaced by a new one (#8) and the MLF is now running at 300 kW and will gradually increase the power to 500 kW.

In the neutron scattering facility there are 23 beam holes, and 20 instruments are under operation to open to general users to investigate structure and dynamics of materials in molecular level, and 1 instrument is under commissioning. World-class scientific outputs have been already created in various scientific fields, ranging from Li-battery science to biomolecular science. Since J-PARC is internationally open for users, we have got experimental proposals from abroad about 15% of the whole proposals. About 30% of proposals have come from industries and a half of them are proprietary use. In the presentation we will show our current status of the neutron target, neutron instruments, sample environments, some statistics in the MLF and new outcomes in science and innovation.

HANARO neutron science facility getting ready for normal operation

J. M. Sungil Park

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Korea*

After a 3 and a half year hiatus, HANARO finally received permission to restart. As of writing, the reactor operators are testing the reactor for safe operation. Meanwhile, neutron scattering and imaging instruments are getting ready for normal operation. Because of lack of funding and personnel, user operation at full capacity will take quite a while.

For the next few years, we plan to open four cold neutron instruments to users: Bio-REF, DC-ToF, Cold-TAS, and G-TS. The first three are the remainders of the cold-neutron project and require finishing touches. The G-TS received an overhaul during the shutdown-period so that it can now do polarized neutron analysis.

Magnon-phonon coupling of hexagonal manganites probed by inelastic neutron and X-ray scattering

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08826, Korea*

Department of Physics & Astronomy, Seoul National University, Seoul 08826, Korea

Magnon and phonon are fundamental quasiparticles of magnetic systems and as such they are supposed to be stable. This well-received wisdom only works up to a point, in which there is neither nonlinear coupling nor cross coupling between the two. Although this high-order term can in principle be present in all real materials, observations of such effects have been rarely made. In this talk, we will explain how this conventional view can break down for hexagonal manganites with specific experimental evidence taken by inelastic neutron and x-ray scattering.

Kitaev quantum spin liquid and its beyond

J. Han^{a,b}, S.-H. Do^{b,c}, H. Gong^{a,b}, B. Kim^{a,b}, K. Kim^b, K.-Y Choi^c, J.-H. Park^{a,b},
K.-S. Kim^b and S. Ji^{a,b}

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37673, Korea*

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The Kitaev quantum spin liquid (QSL) model has fascinated condensed matter physicists for a decade because they are exactly solvable, provide a variety of QSL phases, and are relevant for transition metal compounds such as irridates and ruthenates. This model implements the exchange frustration by bond-dependent Ising-like spin interaction on the ideal two-dimensional honeycomb lattice resulting in an exactly solvable topological QSL ground state and fractional spin excitations represented by noble Majorana fermions. In the past decade, experimental realization of the Kitaev QSL model has been eagerly pursued and now it becomes a challenging to explore exotic quantum phases appearing in real materials due to additional perturbative spin interactions. In this talk, I will introduce recent issues and our findings on α -RuCl₃, a proximate material to the Kitaev QSL.

Small angle neutron scattering study on magnetic skyrmions under various external stimuli

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Y. Yamasaki ^f, J. Suzuki ^b, K. Kakurai ^{a,b}, A. Kikkawa ^a, Y. Taguchi ^a, Y. Tokura ^{a,d},
Y. Iwasa ^{a,d}, and T. Arima ^{a,g}

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^e*J-PARC Center, Japan Atomic Energy Agency, Ibaraki 319-1195, Japan.*

^f*National Institute for Materials Science, Ibaraki 305-0047, Japan.*

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A nanometric vortex-like spin structure ‘magnetic skyrmion’ has attracted increasing attention in recent condensed matter physics research, because topology of the swirling spin texture accounts for a variety of emergent phenomena [1,2]. The magnetic skyrmions were discovered in a metallic chiral magnet MnSi by means of small angle neutron scattering (SANS) experiments [1]; Muhlbauer *et al.* observed a hexagonal SANS pattern indicating that the skyrmions are arranged to form a triangular lattice on a plane perpendicular to applied magnetic field. Subsequent studies have demonstrated that the skyrmions are quite sensitive to various external stimuli, for instance electric current, anisotropic stress, thermal gradient and so on [2]. For recent years, we have been working on SANS measurements on MnSi at TAIKAN instrument (BL15) of MLF in J-PARC. In this talk, we will present our recent studies on stability of skyrmions in MnSi under applications of uniaxial stress [3] and pulsed electric current. In the former study, we observed uniaxial-stress-induced phase transition from the skyrmion lattice phase to a conical magnetic phase. In the latter, we succeeded in observing temporal change of SANS patterns under current pulse application with the time resolution of about 10 ms, by means of stroboscopic SANS technique.

[1] S. Muhlbauer *et al.*, Science **323**, 915 (2009).

[2] N. Nagaosa and Y. Tokura, Nat. Nanotech. **8**, 899 (2013).

[3] Y. Nii, T. Nakajima *et al.* Nat. Commun. **6**, 8539 (2015).

Researches of magnetism and strongly correlated electron systems using the time-of-flight spectrometer 4SEASONS

R. Kajimoto

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4SEASONS (aka *SIKI*) is a time-of-flight direct geometry chopper spectrometer in the Materials and Life Science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC). It is designed for measurements of dynamics in the 10^0 - 10^2 meV energy range [1]. The momentum-energy region for this spectrometer occupies the middle of the momentum-energy space covered by all MLF spectrometers [2]. 4SEASONS was originally designed to meet the requirements for studies of high- T_c oxide superconductors. Indeed, the scientific research activities have been led by studies on copper oxide superconductors, which were followed by studies on iron-based superconductors. In addition, other magnetic materials such as frustrated magnets and multiferroics are also important research themes on 4SEASONS. Including these materials, the field of magnetism and strongly correlated electron systems dominate 80% of the experiment proposals. In the present talk, I will give a brief introduction about the instrument and show recent examples of scientific outputs in this research field.

[1] R. Kajimoto *et al.*, J. Phys. Soc. Jpn. 80, SB025 (2011).

[2] H. Seto *et al.*, Biochim. Biophys. Acta, Gen. Subj. 1861, 3651 (2017).

Magnetic Excitations of the Cu^{2+} Quantum Spin Chain in $\text{Sr}_3\text{CuPtO}_6$

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⁶*School of Mathematics and Physics, The University of Queensland, Australia*

⁷*School of Physics, Georgia Institute of Technology, Atlanta, USA*

⁸*Department of Chemistry, North Carolina State University, Raleigh, USA*

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We report the magnetic excitation spectrum as measured by inelastic neutron scattering for a polycrystalline sample of $\text{Sr}_3\text{CuPtO}_6$. Modeling the data by the 2+4 spinon contributions to the dynamical susceptibility within the chains and with interchain coupling treated in the random phase approximation accounts for the major features of the powder averaged structure factor. The magnetic excitations broaden considerably as temperature is raised, persisting up to above 100 K and displaying a broad transition as previously seen in the susceptibility data. No spin gap is observed in the dispersive spin excitations at low momentum transfer, which is consistent with the gapless spinon continuum expected from the coordinate Bethe ansatz. However, the temperature dependence of the excitation spectrum gives evidence of some very weak antiferromagnetic interchain coupling.

High-energy magnetic excitation of itinerant-electron antiferromagnet Cr studied on HRC and SIKI

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Spin-density waves in Cr are one of interesting research topics in solid-state physics [1]. In spite of its simple *b.c.c.* structure, the magnetism is complicated and still full of many mysteries. One of distinct features is the magnetic excitation that rises with a steep dispersion slope from the incommensurate magnetic Bragg point. The incommensurate magnetic excitation switches to a commensurate one at $\omega \sim 50$ meV, and extends to $\omega > 100$ meV [2, 3]. So far, magnetic scattering was observed in the high energy region of 350 ~ 600 meV using single crystals of Cr on HET at ISIS more than 20 years ago [4]. However, the error of the Q -integrated intensity is large and no information about Q is available.

Recently, we measured magnetic scattering of Cr up to $\omega \sim 250$ meV on a chopper-type spectrometer HRC at J-PARC. The observed magnetic scattering localizes in a narrow Q space at the commensurate position even at $\omega \sim 250$ meV. In this presentation, the ω dependence of line width and integrated intensity is reported. We compare the dispersion relation with theoretical calculations [5] and discuss the energy gap of spin-density waves in Cr.

[1] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).

[2] T. Fukuda *et al.*, J. Phys. Soc. Jpn. **65**, 1418 (1996).

[3] Y. Endoh and P. Böni, J. Phys. Soc. Jpn. **75**, 111002 (2006).

[4] J. R. Lowden *et al.*, J. Mag. Mag. Mater. **140-144**, 1971 (1995).

[5] K. Sugimoto *et al.*, Phys. Rev. B **87**, 134418 (2013).

Magnetic Anisotropy and Magnetoelectric Effect with Spin-Nematic Interaction

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Ba₂CoGe₂O₇ with the noncentrosymmetric crystal structure shows a collinear antiferromagnetic structure below $T_N=6.7$ K. In the magnetically ordered state, a ferroelectric polarization is observed under a magnetic field, and multiferroic property is explained by the spin-dependent d - p hybridization mechanism. In the present study, the neutron scattering measurements were carried out in Ba₂CoGe₂O₇. We found one acoustic and two optical modes in zero field, which are reasonably reproduced by the extended spin wave theory [1]. Furthermore, our result suggests that the anisotropy of the magnetic moments is induced by the spin-nematic interaction. The spin-nematic interaction, which is equivalent to an interaction of local electric polarizations, is the only source of in-plane anisotropy. By using a polarized-neutron-diffraction technique we found that the local-magnetic moment can be controlled by the electric field [2]. The electric-field dependence of the magnetic intensities is consistent with the model that the direction of the magnetic moment rotates from $\langle 100 \rangle$ to $[110]$ with the field. The rotation angle is also explained by spin Hamiltonian including the spin-nematic interaction. The present study revealed the novel magnetoelectric effect that the direction of the local-magnetic moment is controlled continuously by the electric field.

[1] M. Soda *et al.*, Phys. Rev. Lett. **112**, 127205 (2014).

[2] M. Soda *et al.*, Phys. Rev. B **94**, 094418 (2016)

Self-Assembly of Two-dimensional Layered Perovskite Thin Films

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Two-dimensional layered organic-inorganic halide perovskites offer a wide variety of novel functionality such as solar cell and optoelectronics and magnetism. Self-assembly of these materials using solution process (ex. spin coating) makes crystalline thin films synthesized at ambient environment. However, flexibility of organic layer also poses a structure stability issue in perovskite thin films against environment factors (ex. moisture). We have investigated the effect of different polar solvents (water, methanol, a mixture of methanol and water) and relative humidity (RH 5~50%) on self-assembled $(\text{C}_6\text{H}_5\text{CH}_2\text{CH}_2\text{NH}_3)_2(\text{Cu}, \text{Mn})\text{Cl}_4$, (shortly, Cu-PEA, Mn-PEA) on Si substrate by spin coating technique. A combination of X-ray diffraction (XRD) and X-ray absorption spectroscopy (XAS) show that RH has a profound effect on perovskite thin films during sample synthesis and storage, depending on the kind of solvent. The samples prepared using water solvent show quite different from the other cases. According to time-dependent XRD, reversible crystalline-amorphous structural transition takes place depending on RH in the former cases, whereas the latter cases relatively remain stable. It also turns out from XAS that Mn-PEA thin films prepared with solvents such as MeOH, a mixture of MeOH and H_2O , are disordered to the depth of about 4 nm from the surface. Our experimental results highlight that water molecules in atmospheric environment are likely to permeate into bulk along the pathway of organic PEA sublayers and are unlikely to penetrate the inorganic layer along the thickness direction. We suggest that flat and continuous formation is a key factor in the structure stability of two-dimensional organic-inorganic perovskite films against ambient humidity.

Thermal phase stability studies on layered cathode materials for Li-ion batteries using combined *in-situ* high temperature neutron diffraction and gas Analyses

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Li-ion batteries (LIBs) have been considered as the most efficient energy storage systems (ESSs) for electric vehicles (EVs) and grid-scale ESSs owing to their high energy and power densities; however, as the size of battery packs is increasing, there is a growing concern on battery safety issues. Battery safety is closely related to the thermal stability of the cathode materials because the structural transition or decomposition of a charged cathode generally involves O_2 evolution, which increases the risk of a thermal explosion of battery packs.

In this presentation, we will demonstrate the effect of the transition of metal ions in commercial Ni-Co-Mn-based oxide (NCM) electrode materials upon the phase transition at high temperature using combined *in-situ* high-temperature neutron diffraction and gas analyses. We believe that this research can provide a new insight into atomic migration and phase evolution in NCM materials and intuition for the design of cathode materials with high thermal stability.

[1] K.-W. Nam et al. *Adv. Funct. Mater.* **23**, 1047-1063 (2013)

[2] S.-M. Bak et al. *Chem. Mater.* **25**, 337-351 (2013).

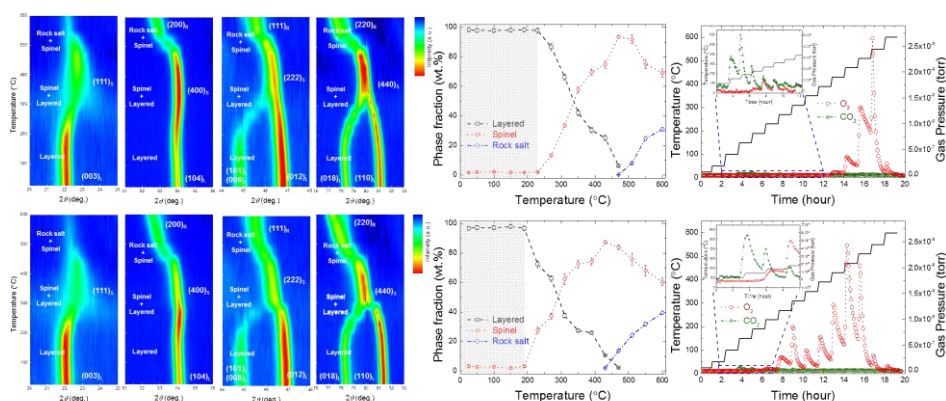


Figure 1. *In-situ* high temperature neutron diffraction patterns, phase fraction and gas analysis data

Multiaxial flow stress measurement based on X-ray diffraction and uncertainty estimation by incorporating crystal plasticity to Monte Carlo method

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^b*National Institute of Standards and Technology, Gaithersburg, MD 20899, USA*

Plastic flow behavior of commercial sheet metal during forming operations is described on the basis of an assumed constitutive model. The uniaxial tension test is a simple method designed to obtain various mechanical properties, with which the accuracy of a constitutive model can be estimated. For instance, in the sheet metal forming community, three angles with respect to rolling direction (i.e., 0°, 45°, and 90°) are typically used in order to examine the mechanical anisotropy. Additional mechanical properties including yield stress, deformation anisotropy, elastic modulus and ductility can be obtained from uniaxial tension tests as well. However, in actual forming operations the loading condition may significantly diverge from that of uniaxial tension, thus requiring experimental verification of the multiaxial and non-linear loading behavior.

In this talk, recent research activities to develop an advanced stress measurement technique using X-ray diffraction [1] will be reviewed. The measurement technique has been successfully applied for an interstitial-free steel sample during large plastic deformation [2]. Moreover, an elasto-viscoplastic self-consistent crystal plasticity model [3] was used to conduct Monte Carlo experiments in order to quantify the uncertainty pertaining to this technique [4].

- [1] T. Foecke, M. A. Iadicola, A. Lin, S. W. Banovic, *Metallurgical and Materials Transaction A* 38, p307-313 (2007)
- [2] Y. Jeong, M. A. Iadicola, T. Gnaeupel-Herold, A. Creuziger, *Acta Materialia* 112, p84-93 (2016)
- [3] H. Wang, P. D. Wu, C. N. Tomé, *Journal of the Mechanics and Physics of Solids* 58, p594-612 (2010)
- [4] Y. Jeong, T. Gnaeupel-Herold, M. Iadicola, A. Creuziger, *Journal of Applied Crystallography* 49, p1991-2004 (2016)

Microstructure and Mechanical Properties of Additive Manufactured Stainless Steel

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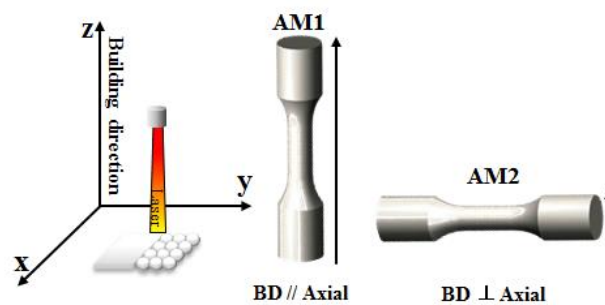
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Additive manufacturing (3D printing) technology has been recently employed for application in a variety of industrial area, such as automobile, aerospace, and medical device due to the advantages of the fabrication of rapid prototype. In this work, two types of specimens are prepared: 1) a cylindrical dog-bone specimen, in which the direction of powder building is perpendicular to the axial loading direction; 2) a cylindrical dog-bone specimen, in which the direction of powder building is parallel to the axial loading direction. In-situ neutron diffraction experiment was performed to investigate the influence of manufacturing direction on uniaxial deformation behavior of 3D printed stainless steel. Lattice strains, diffraction peak intensity, and full width at half maximum (FWHM) of various grain families were measured to understand the deformation behavior at the microstructural level. Strain partitioning and phase evolution between body-centered cubic and face-centered cubic were quantitatively investigated as a function of applied stress. The variations of lattice parameter, phase fraction changes, and strain hardening mechanisms are discussed.



A 3D printed stainless steel with two different manufacturing directions.

Industrial Use of Pulsed Neutron Beam Lines Established by Ibaraki Prefecture in J-PARC MLF.

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Two pulsed neutron beam lines, BL03(iBIX) and BL20(iMATERIA) in J-PARC MLF have been established for industrial use by the local government of Ibaraki prefecture and operated by Ibaraki University. iBIX is for single crystal diffractometry for protein crystal structure analyses, while iMATERIA is for powder diffractometry, small angle scattering and total scattering. The latter beam line has been particularly contributing to the industrial use of J-PARC MLF. The industrial applications amount to as much as about 30% of the total applications in the general user program in J-PARC MLF. The recent activities of these beam lines as well as the policy of the prefecture to promote the industrial use of the pulsed neutron beam lines will be described.

Novel usage of neutrons

White neutron holography for investigations of local atomic structures

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Physical properties of materials can be controlled by impurity doping. This means that distorted atomic structures which appear only around the dopants (local structures) play important roles. The remarkable technique to observe the local structures is atomic resolution holography, which has following advantages: (a) 3D local structures without translation symmetry can be directly observed, (b) Observable range is ~ 20 Å, (c) the central atom can be selected. We are developing white neutron holography at J-PARC, and have succeeded in visualising atomic images around Eu in 1 % Eu doped in CaF_2 , which is a typical scintillation crystal[1]. We are trying to visualise the local structures around dopants in 0.26 % ^{10}B doped Si, 2 % Sm doped LaB_6 , in which characteristic valence behaviour is observed, and 0.06 % B doped 6H-SiC, which is a LED material for white light. Fig.1 shows an atomic image around B in B doped Si. The recent results indicate that white neutron holography can be a direct probe to clarify effects of doping to atomic structures.

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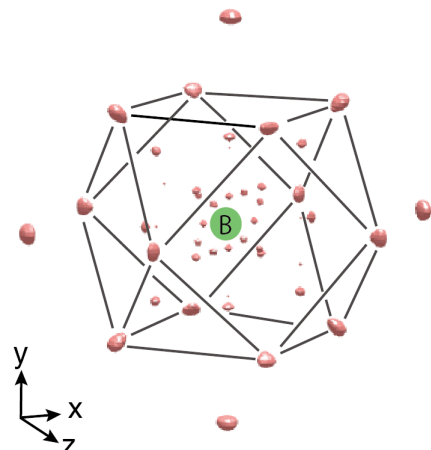


Fig.1 Local atomic image around B of B doped Si

Development of a magnetic imaging technique using pulsed polarized neutrons at J-PARC MLF

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We have been developing a magnetic imaging technique using pulsed polarized neutrons at the Materials and Life Science Experimental Facility (MLF) of J-PARC [1]. In this method, we visualize magnetic field distributions in free space or within a material using wavelength-resolved polarization distribution images. The neutron spin experiences Larmor precession in a magnetic field, and its precession angle depends on the neutron wavelength and field strength integrated along the neutron flight path. Since a change of the neutron spin direction can be detected as a change in the polarization degree, the polarization image at a specified wavelength gives the distribution of the magnetic field along the neutron beam trajectory. Moreover, the integrated field strength at each position in the image can be quantitatively evaluated by analyzing the wavelength dependence of the polarization degree.

To apply this method to the direct observation of the magnetic field of an operating industrial product, e.g. electric motors, electric transformers, etc., we constructed the polarization analysis system for neutron imaging [2] and performed some application studies [3] at BL10 NOBORU and BL22 RADEN. In this presentation, we will describe the present status of our polarized neutron imaging method and show some experimental results, including the imaging of the magnetic field of a rotating electric motor, visualization of the leaked magnetic field from a small electric transformer and observation of the magnetic domains in a grain-oriented electric steel.

This work was supported by Photon and Quantum Basic Research Coordinated Development Program from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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A trial of observation of quantum phase on HRC

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Recently, quantum phase has been recognized as one of physical degrees of freedom, such as spin, charge, orbital, lattice, etc. The fluctuations of these physical degrees of freedom can be observed as spin waves, orbital waves, phonons, and so on, with inelastic neutron scattering. A metallic ferromagnet SrRuO_3 shows band crossing due to spin orbit interactions. The band crossing produces a quantum phase named a Berry phase [1] and behaves as a magnetic monopole in the momentum space, then causes an anomalous Hall effect. The anomalous Hall effect in SrRuO_3 is well described by this picture of the Berry phase [2]. We performed inelastic neutron scattering experiments on SrRuO_3 using the High Resolution Chopper Spectrometer (HRC) at J-PARC, and the temperature dependence of spin waves was investigated. We found that the gap energy of spin waves shows nonmonotonous temperature dependence and that it is well described as a function of the anomalous Hall conductivity [3]. This shows that the fictitious magnetic field of monopole produced by the Berry phase is an observable of inelastic neutron scattering. Spin chirality is also discussed in terms of the Berry phase. In $\text{Nd}_2\text{Mo}_2\text{O}_7$, Mo spins show a noncolinear ordering due to the ordering of Nd spins, then the spin chirality is produced. It is reported that the anomalous Hall effect corresponding to the spin chirality is observed [4]. On the other hand, it is reported that the anomalous Hall effect in $\text{Nd}_2\text{Mo}_2\text{O}_7$ is described by the magnetic moments of Mo and Nd not by the spin chirality [5]. We performed inelastic neutron scattering experiments on $\text{Nd}_2\text{Mo}_2\text{O}_7$, and detected spin waves. These activities using HRC are presented.

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Strain-induced uniaxial magnetic anisotropy variation in epitaxial CoFe₂O₄ thin films[†]

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Magnetic anisotropy is important to significant technological application, such as magnetic and magneto-optic recording media. The high anisotropies have been interested by technological demands such as increasing the magnetic application. Especially, CoFe₂O₄ (CFO) has attracted much interest because of its unique magnetic properties such as larger magnetic elastic constant ($K_1 \sim 2.0 \times 10^6$ erg/cm³ in <100>), large magnetic anisotropy constant ($\lambda_s^p \sim -110 \times 10^{-6}$), excellent chemical stability and high Curie temperature. Furthermore, it has been suggested that these properties can be tuned by the epitaxial strain [1]. In this presentation, we examined the effect of epitaxial strain dependent magnetic anisotropy of epitaxial CFO thin films grown on SrTiO₃ (001) and MgO (001) substrates. Using structural and chemical analysis, we were able to find a correlation between interfacial strain and chemical state vibrations (i.e., cation distributions). Furthermore, the room temperature magnetization measurement also showed a decreased in-plane (out-of-plane) magnetic anisotropy for the films grown on SrTiO₃ (MgO) substrates when epitaxial strain was decreased. The calculated uniaxial magnetic anisotropy energy, based on the experimental data, showed a direct correlation between the uniaxial magnetic anisotropy and degree of epitaxial strain of the films.

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Atomically Engineered Oxide Layers for Novel Magnetoelectric Phenomena

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Materials with simultaneous electric and magnetic ground states show tremendous promise for use in future electronic devices in which electric fields control magnetism. With the technological advancement of thin film synthesis and characterization, several magnetoelectric materials and their coupling mechanisms have been discovered, but with the limitation of weak antiferromagnetism or weak coupling or working at well-below room temperature. Synthesized by reactive molecular-beam epitaxy, perovskite oxide EuTiO_3 that are neither ferroelectric nor ferromagnetic in bulk form, can be transformed into ferroelectric, ferromagnetic, or multiferroic thin films by the misfit strain from the substrates.[1] Atomically engineered oxide $(\text{LuFeO}_3)_m/(\text{LaFe}_2\text{O}_4)$ thin films consisting of LuFeO_3 matrix with additional FeO layers can show magnetically ordered ferroelectricity near room temperature.[2] In this talk, the magnetic structure of these oxides studied by polarized neutron reflectivity and neutron diffraction will be presented.[3]

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Spin-Orbital Correlated Dynamics in the Spinel-Type Vanadium Oxide MnV_2O_4

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In the condensed matter physics, the physical phenomena can be described by the creation, annihilation, and motion of quasi-particles such as phonons and magnons. Recently, the dynamical coupling of two or more degrees of freedom, such as an electromagnon, has been extensively studied. In the present study, we focus on the dynamical coupling between spin and orbital degrees of freedom as an example of such a coupled quasi-particle. We investigated the magnetic dynamics in MnV_2O_4 by inelastic neutron scattering (INS) for observing the spin-orbital coupled waves. The d orbitals of the V^{3+} -site ion, coordinated by an oxygen octahedron, are split into lower-lying t_{2g} orbitals and higher-lying e_g orbitals. During cooling, a phase transition takes place into a collinear ferromagnetic order at $T_C = 58$ K. Below $T_{OO} = 53$ K, the magnetic structure changes into a non-coplanar ferrimagnetic order. Simultaneously, a structural phase transition takes place from cubic to tetragonal, accompanied by the V^{3+} -site orbital order, in which $|yz\rangle$ and $|zx\rangle$ orbitals are alternately arranged along the c -axis. The INS experiment at 5 K shows the presence of spin-wave modes in the energy range between 10 and 17 meV, which was not observed in the previous report [1]. In order to identify spin-wave (SW) modes, we performed SW calculations based on the Heisenberg-type spin Hamiltonian with magnetic anisotropy. We also performed lattice-wave (LW) calculations based on the shell model. The magnetic scattering below 17 meV was sufficiently well reproduced by the SW calculation. In contrast, the scattering around 22 meV, which was considered as SW modes in the previous studies [2], was not be assigned to simple SW and LW modes. We proposed that the 22meV- scattering could originate from the spin-orbital coupled excitation.

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Pressure-induced Quantum Phase Transition in Singlet Ground State Magnet CsFeCl_3 Investigated by Neutron Scattering

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In $S = 1$ antiferromagnet having an easy-plane single-ion anisotropy a quantum phase transition is caused by the competition between the single-ion anisotropy and spin interaction. In the vicinity of the quantum critical point an enhanced longitudinal mode in addition to transverse mode are predicted [1]. An easy-plane type antiferromagnet CsFeCl_3 is an ideal compound for the study; the low-energy excitation of the Fe^{2+} ion is described by a pseudo-spin $s = 1$, and the singlet ground state is realized by the strong easy-plane anisotropy [2]. Furthermore, recent study on the magnetic susceptibility demonstrated that applying pressure induced magnetic long-range order at low temperatures [3].

In order to study the quantum criticality in CsFeCl_3 , we performed both neutron diffraction and inelastic neutron scattering experiments under pressures. The diffraction experiments evidence that a 120° structure is realized in the ordered phase, and the universality class is $U(1) \times Z_2$. Inelastic neutron scattering spectrum exhibits a softening of the low-lying excitation in the disordered phase by applying pressures. In the ordered phase, a well-defined gapped excitation is observed in addition to a continuum-like gapless excitation. Careful comparison with the calculation based on the bond operator theory reveals that the well-defined excitation corresponds a mixed mode of the transverse and longitudinal fluctuations, and the gapless excitation is a transverse mode.

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Development of new 4V-class and zero-strain cathode material for Na ion batteries using neutron diffraction

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We developed $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ as a novel 4V-class and zero-strain cathode material for Na-ion batteries. Through the combined studies using neutron and X-ray diffraction, it was identified that the $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ contains 3-dimensional channels, which enables the facile Na diffusion in the structure. The Na (de)intercalation of $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ was occur at ~ 4 V (vs. Na/Na^+) via the $\text{V}^{3+}/\text{V}^{4+}$ redox reaction and its capacity retention over 3000 cycles was $\sim 67\%$ of the initial capacity. The remarkable cycle stability was due to the near-zero volume change ($\sim 0.24\%$) and the unique centrosymmetric distortion that occurs during a cycle despite the large ionic size of Na ions for (de)intercalation, as demonstrated by *ex-situ* XRD analyses and first-principles calculations. We also demonstrated that the $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ could show the outstanding power capability with $\sim 84\%$ retention of the theoretical capacity at 10C, which is attributed to its intrinsic 3-dimensional open-crystal framework. The combination of this high power capability and extraordinary cycle stability enables the application of $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ to a promising cathode material for Na-ion batteries.

[1]

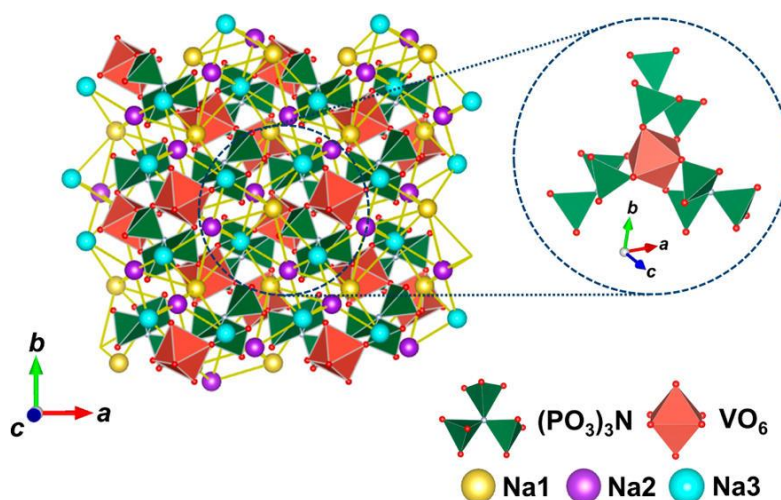


Figure 1. crystal structure of $\text{Na}_3\text{V}(\text{PO}_3)_3\text{N}$ [1]

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Majorana fermions in the Kitaev quantum spin system α -RuCl₃

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Quantum spin liquid (QSL) is a novel disordered ground state without any global symmetry breaking even in zero-temperature. The highly fluctuating ground state by quantum fluctuation emerges the fractionalized quasi-particle excitation in topological order. In particular, the Kitaev QSL [1], arising in bond-directional interacting honeycomb spin-network, is predicted to host Majorana fermions as elementary excitations. By means of a combination of specific heat measurements and inelastic neutron scattering experiments, we demonstrate the emergence of Majorana fermions in single crystals of α -RuCl₃, an experimental realization of the Kitaev spin lattice. The specific heat data unveils a two-stage release of magnetic entropy that is characteristic of localized and itinerant Majorana fermions [2,3]. The neutron scattering results corroborate this picture by revealing quasi-elastic excitations at low energies around the Brillouin zone center and an hour-glass-like magnetic continuum at high energies. Our results confirm the presence of Majorana fermions in the Kitaev quantum spin liquid and provide an opportunity to build a unified conceptual framework for investigating fractionalized excitations in condensed matter.

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Spin wave excitations under strong frustration in the layered kagome compound $\text{CaBaCo}_2\text{Fe}_2\text{O}_7$

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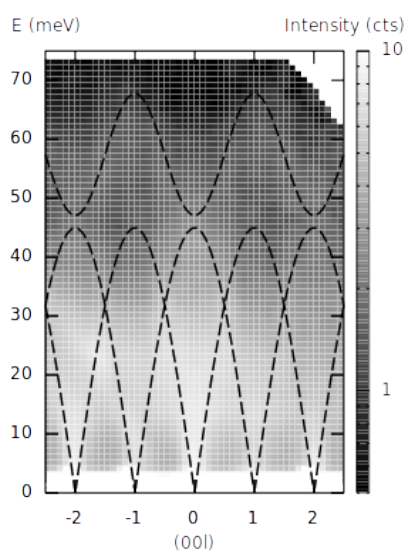
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The layered kagome system in the hexagonal swedenborgite structure [1] displays similarly to the pyrochlores a highly frustrated network of tetrahedral coordinated magnetic ions. Investigated compounds of this family show signs for unusual geometric frustration and disordered ground states despite the typically strong antiferromagnetic (AF) exchange. A Heisenberg nearest neighbor model differentiating between in- and out-of-plane exchange interactions [2] was found to be well suited to describe the magnetic order. The crystallographic structure of the compound $\text{CaBaCo}_2\text{Fe}_2\text{O}_7$ was refined in $P6_3mc$ symmetry ($a=6.36\text{ \AA}$ and $c=10.28\text{ \AA}$) and shows an AF K-type-like ordering below $T_N \approx 160\text{ K}$. [3] Our inelastic neutron scattering



Comparison of observed spin wave excitations with dispersions (lines) extracted from simulations.

study on large single crystals shows strong influences of the peculiar geometric frustration leading to relatively strong damping in the kagome layers, yet, spin wave propagations of larger correlation length perpendicularly (see figure). The experimental results have been discussed in comparison with theoretical calculations and numerical simulations of the spin dynamics. It is found that the excitation spectrum is very sensitive to small changes in order and exchange interactions.

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Structure of Complex Coacervate Core Micelles

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Self-assembled nano particles are usually driven by micro-phase separation between solvent and a part of the molecule, including hydrophobic effect, complexation, and crystallization. In particular, the complex coacervation is a liquid-liquid phase separation when oppositely charged polyelectrolytes are mixed in an aqueous solution. The phenomena has been understood by entropic increase due to counter-ion release and by electrostatic interaction between oppositely charged moieties. Therefore the complex coacervates are stimuli-responsive to pH, salt concentration, polymer concentration, and temperature, which leads to various application such as pharmacy and food industry.

In this study, we investigated the structure of complex coacervate core micelles using block copolyelectrolytes. Two oppositely charged block copolyelectrolytes were prepared by anionic polymerization of poly(ethylene oxide-*b*-allyl glycidyl ether) (PEO-PAGE), and followed by post-modification of the allyl group to introduce functional moieties including sulfonate and ammonium groups. Micelles with coacervate block domains were formed by mixing two AB and A'B diblock copolymer solutions in water where the A and A' blocks are oppositely charged. We observed that coacervation cores are spheres with monodisperse radii and swollen by significant amount of water, and the interface between core and matrix is considerably diffusive with the comparison to the conventional micelles or hydrogels where micro-phase separation is driven by the hydrophobicity of the core-forming blocks. In addition, increasing the salt concentration reduces the coacervation core size and eventually, disintegrate complexation above a certain salt concentration (i.e., critical salt concentration) mainly due by alleviating entropic gain of counter-ion release.

Structure and Mechanical Properties of Elastomer at Surface-modified Carbon Interface

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The interface between polymers and inorganic materials is of pivotal academic and industrial interest, e.g., the interphase in nanocomposites, a class of materials composed of polymers and inorganic nanopowders. In general, the performance of a nanocomposite strongly depends on not only the physical properties of the matrix but also the interaction between the matrix and the filler material. In this study, Structure and mechanical properties of polybutadiene (PB) film on bare or surface modified carbon film were examined. There was the interfacial layer of PB in the vicinity of the carbon layer whose density is higher/lower than that of bulk on the hydrophobic/hydrophilic carbon surface. To extract the information about the structure and the properties of PB at the carbon interface, residual layer (RL) adhering on a carbon surface, which is a model of "bound rubber layer", was obtained by rinsing the PB film by toluene. Density and thickness of RL were identical to that of the the interfacial layer of the PB film. In accordance with change of the density, normal stress was also dependent on the surface free energy; the RLs on hydrophobic carbons were not in rubbery state, presumably in glassy state, whereas that on hydrophilic carbon was in rubbery state. Similarly, wear-test evaluated by atomic force microscopic instruments revealed that the RL on the hydrophilic carbon was peeled off by scratch under the condition of certain stress, while the RLs on the hydrophobic carbons were resistant to the scratch. From these results, we conclude that the surface modification of the carbon affects the interaction between the PB molecules and the carbon surface, and the density and mobility of the PB molecules bound to the surface are altered depending on the strength of the interaction.

Small Angle Scattering Study on Phase Transition of Amphiphilic Molecules in Aqueous Solution

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In aqueous solution, amphiphilic molecules (i. e. surfactants and block copolymers) with both hydrophilic and hydrophobic moieties self-assemble into micelles with various structures and show rich phase behaviors depending on their geometrical molecular shape. Furthermore, the self-assembled structures of amphiphilic molecules can readily transform into other structures under a different external condition such as temperature, concentration, additives and pH, inducing a different geometrical molecular shape. Therefore, amphiphilic molecules have attracted great interests for a wide range of potential applications such as nanotemplates, nanobuilding blocks or drug delivery. However, for practical uses of amphiphilic molecules in nano and bio-technologies, it is required to understand for the phase behaviors of amphiphilic molecules under a variety of external conditions. Therefore, we have investigated the self-assembled structures of amphiphilic molecules (i.e. block copolymers) under the additives and temperature change by using small angle scattering.

Structure analysis of critical gelation cluster on the basis of 3D phase diagram

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Branched polymers on the verge of percolation is called critical polymer clusters. Because of their universal scaling laws, the physics of critical clusters has been well studied in 1970-1990s. In most of these studies, critical polymer clusters have been synthesized by mixing the monomers (or polymers) and the crosslinkers at a critical molar ratio, in which the synthesized branched polymers do not percolate the system after all the reactions ceases. Recently, we established a gelation system by mixing two types of tetra-functional prepolymers, which have complementary reactive end-groups with the other type of prepolymers. Because the reaction conversion is easy to monitor and the side-reactions are almost negligible in this system, we can prepare a series of critical polymer clusters with excellent precision and reproducibility. On the basis of a 3D-phase diagram including reaction conversion, polymer concentration and prepolymer ratio (crosslinker ratio), we measured a series of critical polymer clusters by mixing these two types of prepolymers. With this systematic study, we have confirmed the strong universality of critical polymer clusters in terms of fractal dimension, and for the first time found that the size-distribution is actually a tunable parameter, which has been misunderstood as a universal property (Figure 1) [1].

[1] Li, X.; Hirosawa, K.; Sakai, T.; Gilbert, E. P.; Shibayama, M. SANS Study on Critical Polymer Clusters of Tetra-Functional Polymers. *Macromolecules* 2017, 50 (9), 3655-3661.

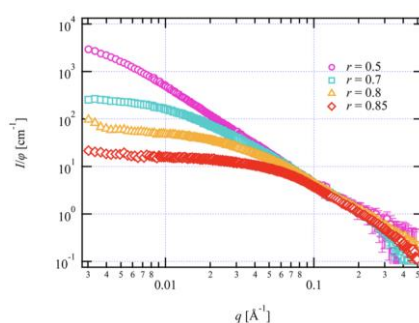


Fig 1. Scattering profile of critical gelation clusters at different critical condition.

Dynamical Behavior of Hydration Water Molecules between Phospholipid Membranes

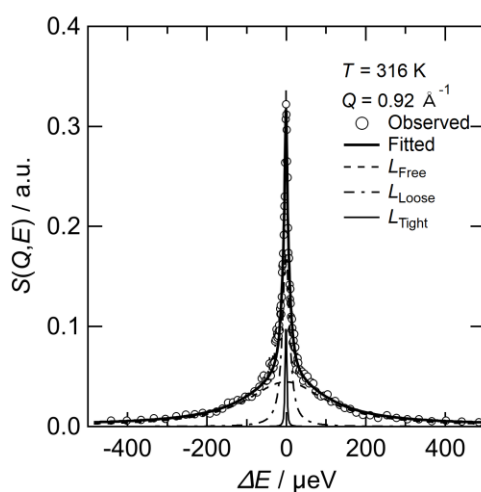
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Biological molecules such as nucleic acids, proteins, and carbohydrates are surrounded by water molecules and their biological functions can only be realized in relation with water. Elucidating dynamical behavior of water near biomolecules is important. In this study, dynamical behavior of hydration water between a lipid bilayer formed by DMPC in sub-nanosecond timescale was investigated by QENS.

The sample used in this study was d_{67} DMPC-37H₂O and DMPC-35D₂O to observe dynamics of the hydration water and DMPC, respectively. QENS experiments were performed at DNA in MLF, J-PARC Tokai, Japan with 3.6 μeV energy resolution. The measured temperature was from 275 to 316 K. The figure shows a QENS profile of d_{67} DMPC-37H₂O at $T = 316$ K and $Q = 0.92 \text{ \AA}^{-1}$. It was well fitted by sum of three Lorentz functions and constant convoluted with the resolution function. The QENS results indicate that the hydration water could be categorized into three types of water; (1) tightly bounded water whose dynamical behavior is cooperative with the DMPC molecule, (2) loosely bounded water whose dynamical behavior is one order of magnitude slower than that of the free water and (3) free water whose dynamical behavior is similar to that of bulk water [1].

[1] T. Yamada, et al., Journal of Physical Chemistry B 121, 8322 (2017).



QENS profile of d_{67} DMPC-37H₂O at $T = 316$ K and $Q = 0.92 \text{ \AA}^{-1}$.

Open circles and the bold solid line are observed intensity and the fitted result, respectively. Other lines are components in the fitting function.

Assembly Architectures of Protein Nanotubes Triggered by Cationic Molecules

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Cells are working by well-designed molecular machines for a variety of tasks: to transfer the genetic blueprint, to move muscles, to transport nutrients, etc. Among them, microtubules (MTs) are involved in dividing cell, intracellular trafficking and defining cell shape. MTs are 25 nm anionic protein nanotubes, comprised of globular dimeric $\alpha\beta$ tubulin subunits aligned end-to-end to form linear protofilaments, which interact laterally to form hollow cylinder. The structures and dynamics of MTs are regulated by molecular switches, such as GTP, chemotherapeutic drug, microtubule-associated-proteins (MAPs), etc. This talk will deal with our recent findings on the assembly architectures of MTs triggered by cationic molecules using small angle X-ray/Neutron scattering and electron micrography.

Neutron reflectivity measurements for diffusion dynamics of polymer melts confined between graphene oxide sheets

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We investigated the diffusion dynamics of entangled polymer thin films confined between graphene oxide (GO) sheets by using neutron reflectivity [1]. We prepared the bilayer of polymethylmethacrylate (PMMA) and deuterated PMMA sandwiched between GO monolayers by employing a Langmuir-Blodgett (LB) and Langmuir-Schaefer (LS) techniques [2]. We found that diffusion coefficient of the PMMA became 30 times slower when the polymers were near the GO surface. This is due to the attractive interaction between the polymer matrix and the GO. The dynamics of polystyrene was reduced only 3 times by the GO confinement, compared to the bulk PS. This is in a good agreement with dewetting results where the dewetting of the PMMA thin films were completely prevented by the GO [3].

- [1] K.-I. Choi, T.-H. Kim, G. Yuan, S. K. Satija, **J. Koo***, *ACS Macro Letters*, 6, 819 (2017).
- [2] H. Kim, Y. R. Jang, J. Yoo, Y.-S. Seo, K.-Y. Kim, J.-S. Lee, S.-D. Park, C.-J. Kim, **J. Koo***, *Langmuir* 30, 2170 (2014)
- [3] T.-H. Kim, H. Kim, K.-I. Choi, J. Yoo, Y.-S. Seo, J.-S. Lee, **J. Koo***, *Langmuir* 32, 12741 (2016).

ABSTRACTS

Poster Presentations

**Magnetic exchange interactions in α -RuCl₃
: Density functional theory study**

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Based on the density functional theory, we study the magnetic exchange interactions of α -RuCl₃, a promising candidate of Kitaev system. By precisely mapping the total energies of various noncollinear spin configurations onto a generalized Kitaev-Heisenberg Hamiltonian, we have obtained various isotropic and anisotropic interaction parameters within a fully *ab initio* scheme. Especially, we discuss the importance of further neighbor anisotropic terms and interlayer interactions in the stabilization of the magnetic order in the system. We compare our approach to the previous studies, and discuss the possible way of tuning the exchange interactions.

Magnetic Structure of a Non-Centrosymmetric CePdSi₃

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Since the discovery of the first heavy electron superconductor CePt₃Si [1] with a non-centrosymmetric crystal structure, the effect of the antisymmetric spin-orbit interaction (ASOI) on superconductivity and magnetism in correlated *f* electron physics has attracted much attention. Here we focus on the non-centrosymmetric BaNiSn₃-type compound CePdSi₃. It exhibits successive magnetic transitions, weak ferromagnetism, and unusually complex metamagnetic transitions [2]. Since this crystal structure does not cause geometric frustration such as a triangular lattice, influence of ASOI may drive multiple metamagnetic transitions.

As the first step of further understanding of the ASOI effect, we examine the magnetic structure of CePdSi₃. By the neutron diffraction experiments using a single crystalline sample, we observed magnetic reflections at $\tau_1 = (0.32, 0, 0)$ in the phase I, and at $\tau_2 = (0.32, 0, 1)$ in addition to $\tau_1 = (0.32, 0, 0)$ in the phase III. We found that the magnetic structure of CePdSi₃ is a longitudinal spin density wave similar to those in CeRhSi₃ and CeIrSi₃.

[1] E. Bauer *et al.*, Phys. Rev. Lett., **92**, 027003 (2004)

[2] D. Ueta, Y. Ikeda and H. Yoshizawa, J. Phys. Soc. Jpn., **85**, 1014703 (2016)

Dynamic critical behavior in Kitaev–Heisenberg model

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Kitaev honeycomb lattice model [1] is a unique example that is exactly solvable and have spin liquid ground state. The spin liquid ground state is topological, highly entangled state, and has abelian or non-abelian anyon excitations. These properties lead the model to be an ideal system for the topological quantum computation applications. However, in real material, such as iridate [2] or ruthenate compounds [3], there are inevitably ordering term such as Heisenberg term, make the ground state ordered. This ordered ground state may be removed by applying external magnetic field and make spin liquid [4]. Here we study Kitaev-Heisenberg model and investigate quantum criticality of the magnetic transition point. We calculate observable properties such as spin susceptibility and show the critical behavior different from Ising universality class due to the interaction with fractionalized particles from Kitaev Hamiltonian.

[1] A.Kitaev, *Annals of Physics* 321, 2 (2006).

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[3] K. W. Plumb et al., *Physical Review B* 90, 041112(R) (2014).

[4] S.-H. Baek *et al.*, *Physical Review Letters* 119, 037201 (2017).

Crystalline Electric Field (CEF) Level scheme of the Non-Centrosymmetric CeTSi₃ (T = Rh, Ir)

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The heavy-electron superconductors CeRhSi₃ and CeIrSi₃ are quite interesting materials to examine the influence of the lack of inversion symmetry in the crystal structure [1][2]. Interestingly, their superconducting upper critical fields exceed both of the Pauli and orbital limiting fields. Such behavior cannot be explained by the conventional BCS theory for the superconductivity, but is attributed to the influence of the antisymmetric spin-orbit interaction (ASOI). We have grown single crystalline samples of CePdSi₃ and CePtSi₃ which have the same BaNiSn₃-type structure (*I4mm*) with the Rh and Ir systems, and carried out magnetization measurements and inelastic neutron scattering (INS) measurements in order to explore the difference of magnetic behavior from those of the Rh and Ir systems [3][4]. One of the different features we noticed is the wave functions of the ground state derived from the CEF level schemes. For the superconducting Rh and Ir systems, their wave functions of the ground state are Γ_7 , whereas for non-superconducting Pd and Pt systems, those are Γ_6 . We found that these wave functions provide qualitatively different ground states.

Furthermore, in a proceeding report of the INS experiment of CeRhSi₃, there observed no clearly resolved peaks which correspond to two CEF excitations [5]. As for the CEF level scheme of CeIrSi₃, it was only estimated from the magnetization measurements [6]. Therefore, we performed the INS experiments on polycrystalline samples of CeRhSi₃ and CeIrSi₃ using the High Resolution Chopper (HRC) spectrometer installed at BL12 in J-PARC MLF, and we tried to directly observe their CEF peaks.

- [1] N. Kimura *et al.*, Phys. Rev. Lett., **95**, 247004 (2005)
- [2] I. Sugitani *et al.*, Jour. Phys. Soc. Jpn., **75**, 043703(2006)
- [3] M. Smidman, PhD. Thesis, Univ. of Warwick UK, (2014)
- [4] D. Ueta *et al.*, J. PhysB. Condensed Matter, (2017).
- [5] N. Kimura *et al.*, J. Phys. Soc. Jpn., **76**, 051010(2007)
- [6] Y. Okuda *et al.*, J. Phys. Soc. Jpn., **76**, 044708(2007)

Lattice symmetry breaking by ionic orbital doping in tetragonal spinel Mn_3O_4

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Mn_3O_4 is a well-known tetragonal ferrimagnetic spinel with Mn^{2+} ($3d^5$, $S = 5/2$) and Mn^{3+} ($3d^4$, $S = 2$) ions on tetrahedral and octahedral sites, respectively. The Jahn-Teller effect of Mn^{3+} ions elongates the cubic lattice along the c axis, which makes the antiferromagnetic nearest-neighbor exchange stronger within the c planes. Recently, orthorhombic lattice instabilities were observed below T_N suggesting the persistent geometrical frustrations between the antiferromagnetic chains crossing orthogonally each other.

Using synchrotron x-ray and neutron diffraction measurements, we investigated how the crystal and magnetic structures of Mn_3O_4 change when metal ions replaced a few percent of Mn^{2+} ions on tetrahedral sites. When Cu^{2+} ($3d^9$) or Co^{2+} ($3d^7$) ions were doped with odd number of electrons in the $3d$ orbital, the orthorhombic phases were stabilized below T_N . This lattice symmetry breaking is attributed to the odd number of electrons in the tetrahedral t_2 orbitals. We also found that the orthorhombic strain increased linearly with the Cu^{2+} doping concentration whereas it remained nearly constant up to 30% of Co^{2+} doping. Such distinct behavior is most likely due to the coexistence between high and low spin states of Co^{2+} ions, which does not provide sufficient internal stress albeit breaking the lattice symmetry. In contrast, such orthogonal distortions were not observed when Ni^{2+} ($3d^8$) ions were doped introducing even number of electrons.

Inelastic Neutron Scattering in Kagome-Triangular Lattice CsCrF₄

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Frustrated magnets have been paid a lot of attention because of their nontrivial magnetic states at low temperatures. One of the examples is the Kagome-Triangular (KT) lattice that has unusual path of the 2nd neighbor interaction in the Kagome lattice as shown in Fig. 1 [1]. CsCrF₄ is a rare experimental realization of the KT lattice where Cr³⁺ ions ($3d^7$) carry spins $S=3/2$. Recent neutron diffraction experiment showed that the 120° structure with the propagation vector of $\mathbf{Q} = (1/2, 0, 1/2)$ appears at low temperatures [2]. The structure had never been predicted in the phase diagram of the Heisenberg KT magnet [1]. From the classical calculation of the ground state, it was found that the nearest-neighbor Kagome interaction, Dzyaloshinskii- Moriya (DM) interaction, and single ion anisotropy play a key role for the appearance of the structure [2].

We performed inelastic neutron scattering experiments to identify the spin model of CsCrF₄ and to reveal the origin of the nontrivial 120° structure. In this experiment, we used High Resolution Chopper spectrometer in J-PARC. The features of the observed magnetic excitations are as follows: the bandwidth of 9 meV, a flat structure at 0.5 meV and 1.5 meV around $Q = 1 \text{ \AA}^{-1}$, and continuous intensities at $6 \text{ meV} < \hbar\omega < 9 \text{ meV}$ in the entire range of measured Q . The comparison between the simulation by the spin wave approximation and the experimental results reveals that the ferromagnetic KT lattices with the antiferromagnetic 2nd neighbor interaction in the crystallographic ab planes are antiferromagnetically coupled in the c direction. Furthermore, the flat structure at 0.5 meV and 1.5 meV is the result of the lift of the ground state by single-ion anisotropy and DM interaction.

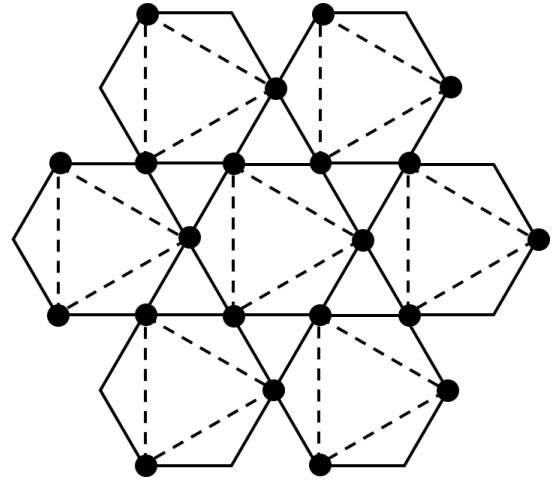


Fig. 1. Schematic description of the Kagome-Triangular lattice. Solid and dashed lines are the first and second neighbor Kagome interactions.

[1] H. Ishikawa *et al.*, J. Phys. Soc. Jpn **83**, 043703 (2014)

[2] M. Hagihala *et al.*, to be published.

Spin dynamics of diluted non-collinear antiferromagnet $\text{YMn}_{1-x}\text{Al}_x\text{O}_3$ ($x = 0, 0.05, 0.1$ and 0.15)

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In condensed matter physics, low-dimensional systems lead to qualitatively new physics due to their unique properties and exotic phenomena that were not found in higher dimensional systems. In such systems, any kinds of weak perturbation can induce new competing ground states and so a novel rich phase diagram [1]. A particularly important case is the spin systems with non-magnetic impurity. Here we report the inelastic neutron scattering (INS) experiments on the hexagonal $\text{YMn}_{1-x}\text{Al}_x\text{O}_3$ ($x = 0, 0.05, 0.15$) single crystals. INS measurements on Al doped single crystals reveal the large broadening of magnon linewidth around B point, as predicted in theoretical study [2]. Spin wave simulation using the spin cluster calculation captures most of details of measured data. This shall discuss the possible interpretation of result in terms of umklapp scattering off the impurity-induced spin texture.

[1] E. Dagotto, Science **309**, 257 (2005).

[2] V. S. Maryasin et al., Phys. Rev. Lett. **111**, 247201 (2013).

Controlling the stoichiometry of triangular lattice antiferromagnet $\text{Li}_{1+x}\text{Zn}_{2-y}\text{Mo}_3\text{O}_8$

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An intriguing topic in condensed matter physics is exploring exotic ground states in frustrated systems where competing interactions destabilize conventional magnetic order, such as the quantum spin liquid and the resonating valence bond state. The compound $\text{LiZn}_2\text{Mo}_3\text{O}_8$ (LZMO) is an antiferromagnet with magnetic Mo_3O_{13} clusters forming triangular layers which is proposed to realize a condensed valence bond state [1]. For the ideal composition, these clusters have an unpaired electron (spin 1/2) and hence the system may be optimal for exploring exotic ground states in $S = 1/2$ triangular lattice. However, intersite disorder of Li and Zn in LZMO easily leads to off-stoichiometry that introduces unoccupied $S = 1/2$ sites. Partial control of the Zn composition has been achieved by earlier efforts involving an electrochemical technique [2], but full stoichiometry control has not been achieved as far as we know.

Since the stoichiometry is the key to explore intrinsic physics in the $S = 1/2$ triangular lattice, we determined the chemical composition ($\text{Li}_{1+x}\text{Zn}_{2-y}\text{Mo}_3\text{O}_8$) of several polycrystalline compounds that were prepared with widely varied starting compositions by modifying the previously reported solid-state reaction technique [1]. Characterization was done by ICP mass spectroscopy, X-ray diffraction and neutron powder diffraction (NPD) at Echidna, ANSTO. Composition dependence on the magnetism was investigated using MPMS magnetometer. Polarized neutron scattering was performed at HB-1 PTAX (ORNL).

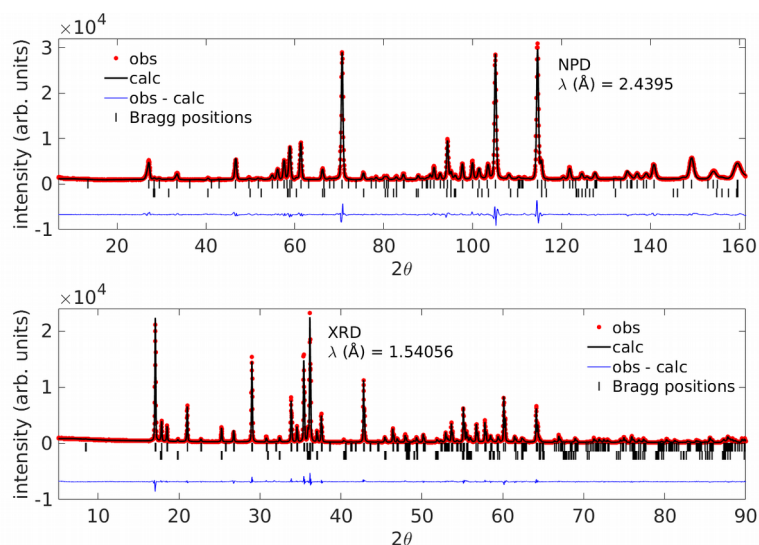


Fig. 1. Combined Rietveld analysis on a improved stoichiometry LZMO sample.

The stoichiometry of Li and Zn was improved, exemplified in Fig. 1; NPD + XRD combined Rietveld analysis confirms that the sample closest to the stoichiometry has $x = -0.06(6)$, $y = 0.086(8)$. The magnetic susceptibility χ of the improved stoichiometry samples shows a broad hump around $T = 160$ K not reported in previous work [1] suggesting that short-range antiferromagnetic order extends up to 100~200K.

[1] J. P. Sheckelton *et al.*, Nat. Mater. 11, 493-496 (2012).

[2] J. P. Sheckelton *et al.*, Mater. Horizons 2, 76-80 (2015).

Weak-ferromagnetism of CoF_3 and FeF_3

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Cobalt trifluoride CoF_3 and Iron trifluoride FeF_3 are known as a G-type antiferromagnetic structure in the $R\text{-}3c$ rhombohedral crystal structure. The neutron powder diffraction study had been reported that Co antiferromagnetic spin direction is parallel to 3-fold crystal axis in CoF_3 whereas Fe antiferromagnetic spin direction is perpendicular to 3-fold crystal axis in FeF_3 . However, the resolution of early neutron powder diffractometer is not enough to distinguish between $(003)_h$ and $(101)_h$ rhombohedral magnetic peaks definitely. We confirm the proposed magnetic structural models by high-resolution powder neutron diffractometer(SuperHRPD) at room temperature in MLF, J-PARC. In addition, we discuss weak-ferromagnetism by antiferromagnetic spin directions and Dzyaloshinski-Moriya interactions.

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Magnetic study of oxygen molecule adsorbed in Nanoporous complex $\text{Cu}_2(4\text{-F-bza})_4(2\text{mpyz})$

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In some types of nanoporous metal organic complex, oxygen molecules are adsorbed in the porous and form the three dimensional periodic structure. One of the most interesting examples is a Cu complex, CPL-1, where the adsorbed O_2 molecules form ladder like structure and $S = 1$ spin gap system [1]. The neutron spectrum at low temperature was explained by the singlet-triplet excitation of $S = 1$ dimer [2]. Meanwhile the temperature dependence deviates from the dimer model. Combination of calculation on spin-dependent molecular potential [3] and neutron scattering results indicated that the origin of the unconventional dynamics was the soft framework of oxygen supercrystal.

A Cu complex, $\text{Cu}_2(4\text{-F-bza})_4(2\text{-mpyz})$, is a new nanoporous material. The adsorbed O_2 molecules form trimer structure [4]. In order to investigate the magnetism of the oxygen molecules adsorbed in $\text{Cu}_2(4\text{-F-bza})_4(2\text{-mpyz})$, we performed the magnetization measurement and the inelastic neutron scattering experiment on cold-neutron TOF spectrometer PELICAN installed at ANSTO. The magnetization approaches to $2 \mu_B / 3\text{O}_2$ in M - H curves measured below 2 K, which indicates the formation of the trimer. We observed non-dispersive excitation at 0.4 meV in the inelastic neutron scattering spectra for O_2 -adsorbed $\text{Cu}_2(4\text{-F-bza})_4(2\text{-mpyz})$ at 4.6 K, which cannot be observed in the spectra for the sample after O_2 gas evacuation. We analyzed the excitation based on the $S = 1$ antiferromagnetic trimer model.

[1] R. Kitaura *et al.*, Science **298**, 2358 (2002). [2] T. Masuda *et al.*, J. Phys. Soc. Jpn. **77**, 083703 (2008). [3] B. Bussery and P.E.S. Wormer, J. Chem. Phys. **99**, 1230 (1993). [4] Y. Takasaki and S. Takamizawa, JACS **136**, 6806 (2014).

Magnetic anisotropy on multiferroics $\text{Ba}_2\text{MnGe}_2\text{O}_7$

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$\text{Ba}_2\text{MnGe}_2\text{O}_7$ is a square-lattice antiferromagnet where an isotropic classical spin $S = 5/2$ is localized at Mn^{2+} ion ($3d^5$). The ground state is a collinear antiferromagnetic order, and the transition temperature T_N is 4K. The spin is aligned in the ab -plane. The magnetic spectrum measured by the triple-axis spectrometer was explained by the square-lattice Heisenberg model which has weak coupling [1]. Below T_N , an electric polarization is simultaneously induced by applying a magnetic field along $[110]$ direction [2]. The microscopic origin of multiferroicity is the spin-dependent d - p hybridization mechanism [3], where the local electric polarization is described by a quadrupole operator of Mn spin. Recently in the isostructural compound $\text{Ba}_2\text{CoGe}_2\text{O}_7$ having the large single-ion anisotropy of easy-plane type, it was founded that the spin quadrupole interaction led to the 4-fold anisotropy in the ab -plane, and the magnitude of the quadrupole interaction represented a controllability of the spin by the electric field [4, 5]. In contrast in $\text{Ba}_2\text{MnGe}_2\text{O}_7$, the spin is isotropic, and the quadrupole interaction is weak as well as the single-ion anisotropy because of the absence of orbital angular momentum of Mn^{2+} ion. Precise measurement of the anisotropy energy is important.

In this study we performed bulk magnetization measurement and inelastic neutron scattering (INS) experiment using the near-backscattering spectrometer DNA installed at J-PARC MLF, the energy resolution of which is as good as 3 μeV . The INS spectrum reproduces the previous one [1], and the anisotropy gaps of 0.037 meV and 0.11 meV at $\mathbf{Q} = (1, 0, 0.5)$ is newly observed. To explain this spectrum, the spin Hamiltonian needs the spin-nematic interaction and the easy-plane anisotropy with the previous Heisenberg model [1]. In addition the magnitude of the anisotropy gap and spin-flop field and antiferromagnetic magnetization yield to a unique function of temperature. This suggests the origin of anisotropy is the spin-nematic interaction.

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[3] T. Arima, J. Phys. Soc. Jpn. **76**, 073702 (2007). [4] M. Soda *et al.*, Phys. Rev. B. **94**, 094418 (2016).

[5] M. Soda *et al.*, Phys. Rev. Lett. **112**, 127205 (2014).

Correlation between gas transport properties and the morphology/dynamics of crystalline fluorinated copolymer film

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The crystalline structure, dynamics, and gas transport properties (i.e., the gas permeability, gas diffusion coefficient, and gas solubility coefficient) of poly(tetrafluoroethylene-co-perfluoroethylvinylether) (PFA) membranes were systematically investigated via differential scanning calorimetry, wide/small/ultra-small-angle X-ray scattering, and quasielastic neutron scattering measurements. We evaluated the gas transport properties using a constant-volume/variable-pressure method. The gas permeability and the gas diffusion coefficient decreased with increasing crystallinity of the PFA membranes at crystallinities below 32%. However, in membranes with a crystallinity of 32% or greater, these parameters depended on the characteristics of the gas molecules, such as their kinetic diameter. The so-called long spacing period and the thickness of the crystalline/amorphous regions increased with crystallinity according to the small/ultra-small-angle X-ray scattering results. Furthermore, the quasielastic neutron scattering measurements indicated that the scattering law was well fitted to a sum of narrow and broad Lorentzian components. In particular, the narrow components, i.e., the local motion of amorphous components and side chains, increased with crystallinity. These results suggest that large gas molecules could pass through the PFA membranes, assisted by the motion in the amorphous region. [1]

[1] T Nyuui, G Matsuba, S Sato, K Nagai and A Fujimori, *J. Appl. Polym. Sci.*, **135**(2), 45565 (2018).

The phase transitions of micellar structure in dilute polymer-polymer system

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The amphiphilic block copolymers self-assemble into a nanoscale micellar structure in solution, and their phase can be easily changed by their surrounding conditions or its mass fraction of hydrophilic/phobic [1]. Therefore, the micellar structures can be controlled by mass ratio of their consisted chain. The Pluronic triblock copolymer are amphiphilic molecules with poly(ethylene oxide) (PEO) blocks, which are water soluble, and poly(propylene oxide) (PPO) blocks, which are water insoluble at low temperature but water insoluble at ambient temperature, combined into single polymer chain. It is anticipated that when the block copolymer is mixed with another block copolymer, leading to the formation of the block copolymer-block copolymer complex, a new self-assembled structure can be induced without complicated synthesis of the block copolymer with different mass ratios of each block. While the mixture system of two kinds of block copolymers is an easy and simple method, it has not been fully exploited yet. Herein, we report a new approach to control the phase behavior of Pluronic block copolymer (P85, PEO₂₆PPO₄₀PEO₂₆) or (F127, PEO₉₉PPO₆₅PEO₉₉) by adding another Pluronic block copolymer (PE6200, PEO_{10.5}PPO₃₀PEO_{10.5}) that has a different mass fraction of the hydrophilic moiety. Small angle neutron scattering measurements showed that, depending on the adding PE6200 concentration and temperature, the Pluronic P85 or F127 block copolymer with a random coil structure transformed into spheres, cylinders, and vesicles, consistent with the results of visual inspection and dynamic light scattering measurements. This is the first demonstration of a simple method to control the phase behavior of a block copolymer using the same block copolymer but with different mass ratio of each block, affording a convenient means to control the self-assembly

of block copolymers in aqueous solution. The self-assembled structures of block copolymers thus can provide a broad spectrum of potential applications in nano- or bio-science such as nano-building blocks [2] and drug delivery systems [3].

[1] Shaoyong Yu, Tony Azzam, Isabelle Rouiller, and Adi Eisenberg, *J. Am. Chem. Soc.*, 2009, **131** (30), 10557–10566.

[2] Zhenli Zhang, Mark A. Horsch, Monica H. Lamm, and Sharon C. Glotzer, Tethered Nano Building Blocks: Toward a Conceptual Framework for Nanoparticle Self-Assembly, *Nano Letters*, 2003, **3**, 1341-1346.

[3] G. Wanka, H. Hoffmann, and W. Ulbricht, Phase Diagrams and Aggregation Behavior of Poly (oxyethylene)-Poly (oxypropylene) –Poly (oxyethylene) Triblock Copolymers in Aqueous Solutions, *Macromolecules* 1994, **27**, 4145-4159.

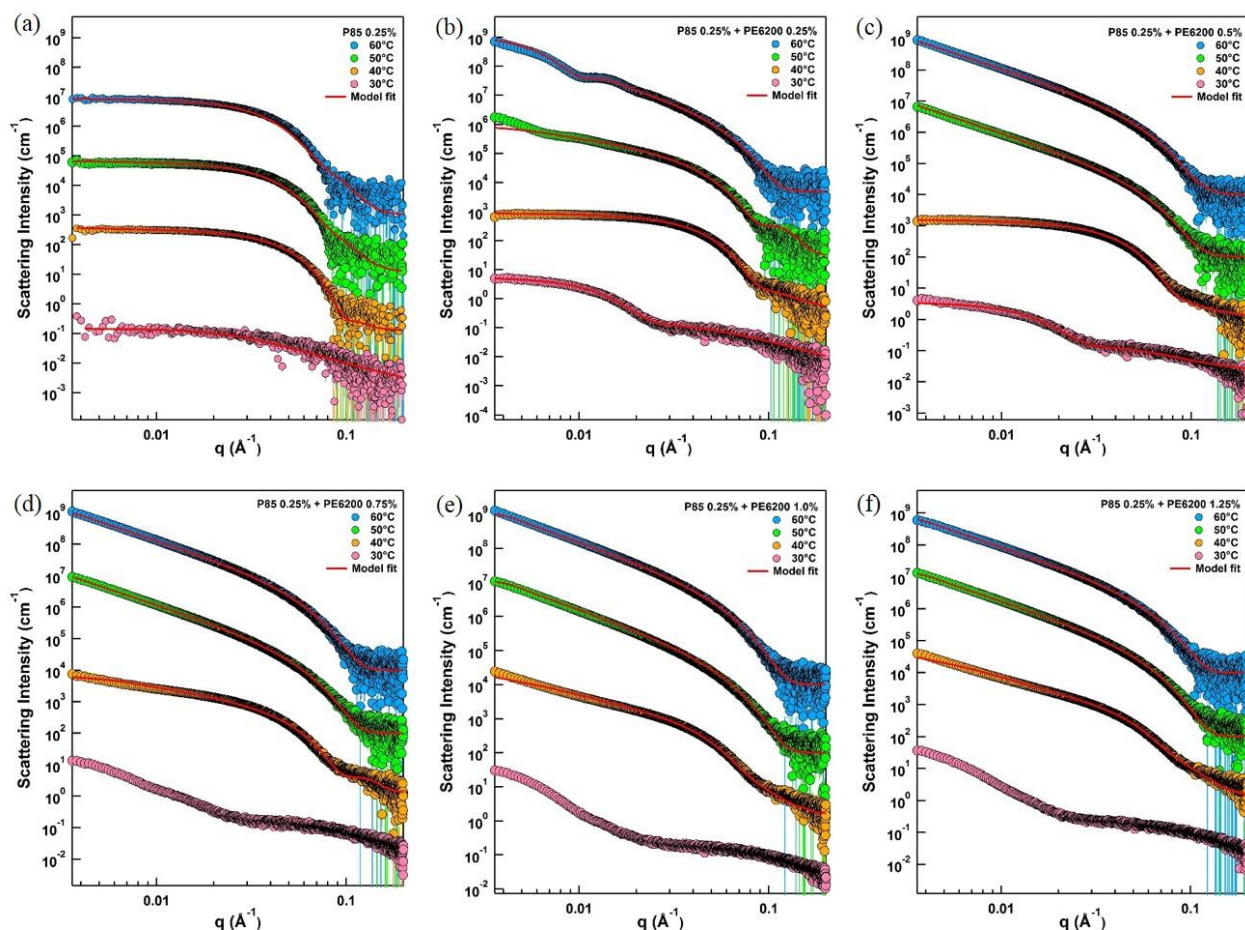


Figure. 1. SANS intensities of the P85-PE6200 mixtures at varying temperatures. SANS intensities of the P85 (0.25%)-PE6200 mixture in water with increasing PE6200 concentrations at (a) 0%, (b) 0.25%, (c) 0.5%, (d) 0.75%, (e) 1.0%, (f) 1.25% when the temperature increases from 30 °C to 60 °C. SANS intensities were vertically shifted for visual clarity.

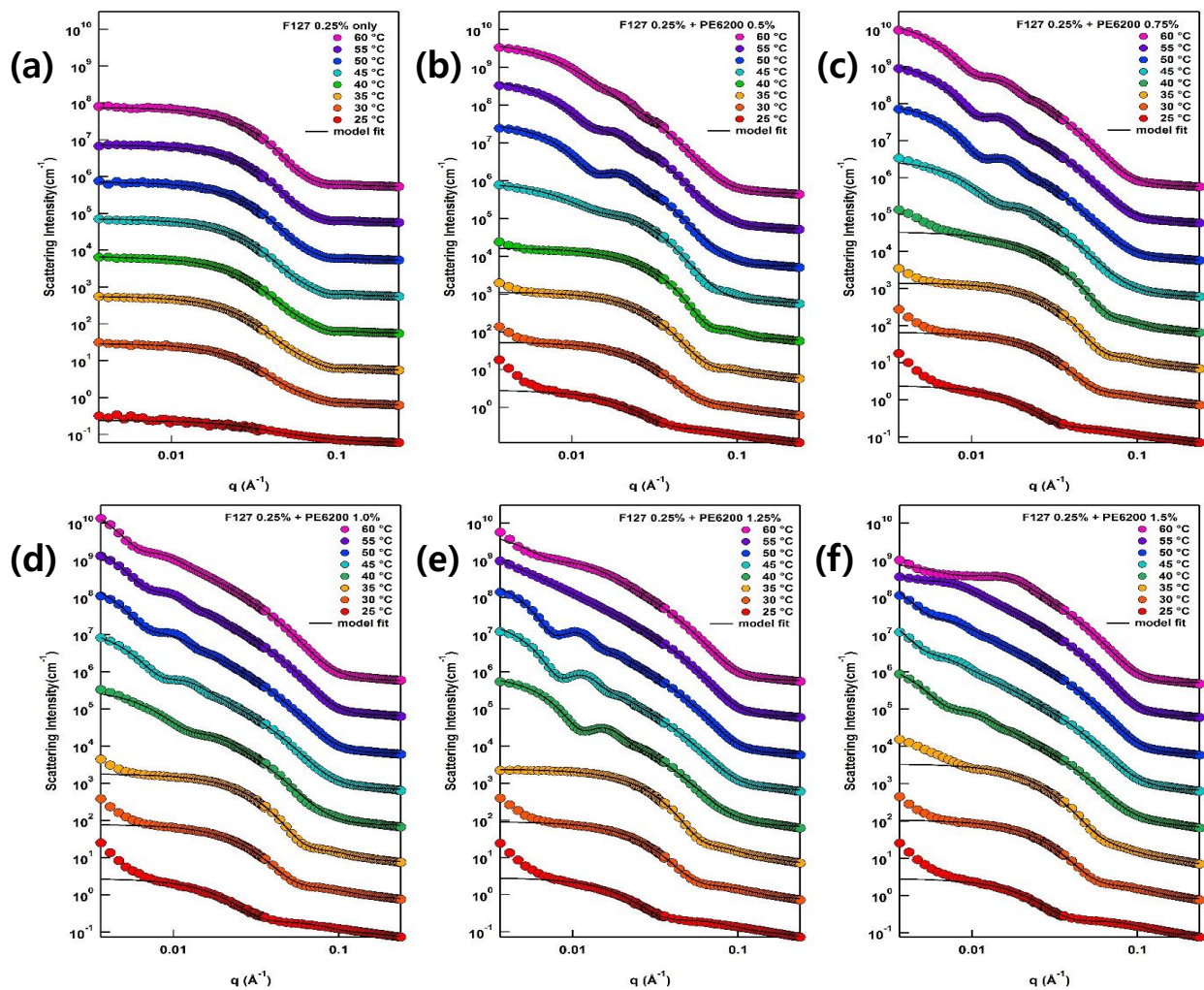


Figure. 2. SANS intensities of the F127-PE6200 mixtures at varying temperatures. SANS intensities of the F127 (0.25%)-PE6200 mixture in water with increasing PE6200 concentrations at (a) 0%, (b) 0.5%, (c) 0.75%, (d) 1.0%, (e) 1.25%, (f) 1.5% when the temperature increases from 25 °C to 60 °C. SANS intensities were vertically shifted for visual clarity.

Microscopic Solvation Structure and Phase Behavior of Thermo-responsive Polymers in Ionic Liquids

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Ionic liquids (ILs) are salts which have their melting temperature below room temperature. Because ILs have high ion atmosphere, they form unique solvation environment for solutes. Recently, it was reported that poly(benzyl methacrylate) (PBnMA) and PBnMA derivatives exhibit a lower critical solution temperature (LCST)-type phase separation in ILs.^{1, 2} One of the unique characteristics of the thermo-responsive polymer in IL systems is that modification on chemical structure of polymers and ILs causes drastic variation of cloud point temperature of the system.² It indicates that microscopic molecular interactions between polymer and IL control the macroscopic phase behavior of the polymer in IL solution.

In this study, we focused on solvated structure of the thermo-responsive polymers in IL solutions. We carried out small angle neutron scattering (SANS) experiments on the PBnMA derivatives in deuterated IL solutions which exhibit the LCST-type phase separation. The effective interaction parameter (χ) between the polymers and ILs was evaluated from the obtained SANS profiles. For all the combination of polymers and ILs, χ increased with temperature, indicating that solvent quality of the ILs becomes worse at elevated temperature. The enthalpic contribution (χ_H) to χ was extracted from the temperature dependence of χ . As a result, χ_H strongly depends on the chemical structures of polymers and ILs. Furthermore, the microscopic solvation structure of the polymers in ILs was investigated by means of high-energy X-ray diffraction (HEXTS) experiments and all-atom molecular dynamics (MD) simulations. Based on the structural analysis with higher spatial resolution, we revealed the spatial distribution of IL ions around polymer chain. It was found that the effect of the chemical modifications on χ_H is ascribed to the microscopic solvation structure.

- [1] T. Ueki and M. Watanabe *Langmuir* **2007**, 23, 988-990.
- [2] K. Kodama *et al.*, *Langmuir* **2009**, 25, 3820-3824.

Effect of graphene oxide sheets on diffusion dynamics of confined polymer thin films

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Carbon-based nanomaterials, such as graphene oxide(GO), have been in interest due to enhancement of polymer's physical properties. The interaction between the GO and polymers significantly affects the polymer dynamics in confined system. In this work, we used polymer thin films sandwiched between GO monolayers as a model composite system to quantitatively measure the mobility change of polymers near GO surfaces. We investigated three different interaction, namely, a strongly attractive, a weakly attractive and a repulsive interaction. Polymer mobility of confined systems was analyzed a neutron reflectivity technique. The diffusion dynamics was compared as a function of polymer bilayer symmetric and asymmetric thicknesses. Consequently, when the film is thin (~ 1 Rg), diffusion coefficient decreases for poly(methyl methacrylate) (PMMA) thin films sandwiched between GO sheets. However, diffusion coefficient increases for PMMA thin films sandwiched between PS thin films. This is due to strongly attractive interaction between the GO and PMMA, but repulsive interaction between PS and PMMA.

Mechanical Property Control of Polymer Nanocomposites Using Dopamine-Modified Brush Polymer at Nanoparticle Surface

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The physical property of polymer nanocomposites (PNCs) relies on the state of particle dispersion in polymer matrix, requiring an effective control of particle dispersion. One of the reliable methods is to change the surface chemistry of particles using chemical grafting with polymers, which often requires multiple synthetic processes.

In this study, we report the particle dispersion in PNCs and its resulting mechanical property can be readily altered via simple physical grafting with dopamine-modified brush polymer. The silica nanopartilces were coated with dopamine-derived poly(ethylene glycol) (DOPA-PEG) and dispersed in PEG matrix with varying molecular weights. The DOPA-PEG adsorbs on silica surface irreversibly, increasing effective size of particles. We found that the DOPA-PEG can change the wetting property with polymer matrix at the interface, which greatly influences on the state of particle dispersion and its mechanical property.

The detailed microstructure and rheological property are studied with small angle x-ray scattering (SAXS) and oscillatory shear experiments. Additional small angle neutron scattering (SANS) experiments will be performed to analyze the detailed microstructure of adsorbed polymer on particle surfaces.

Quantitative evaluation of uniformity of polymer networks by SANS with contrast matching technique

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Polymer gel is a material with flexibility but no flowability, which has many applications in food industry and bio-medical fields. The mechanical properties and swelling behavior of polymer gels depend on the network structure of the polymer gels. However, it is still difficult to “quantitatively” evaluate the homogeneity of the structure of the gels. In a previous study by Benoit et al¹, the average distance between deuterium-labeled crosslinkers were successfully obtained for a polymer gel synthesized by living radical reaction. Following the strategy of Benoit, we have synthesized the tetra-PEG gel with deuterium-label near the branching point and conducted SANS experiments with contrast matching method. However, we failed to observe any peaks (Figure 1). The reason is likely due to the labeled part was too small to scatter enough neutrons.

[1] Benoit, H. et al. Journal of polymer science 1976, 14, 2119.

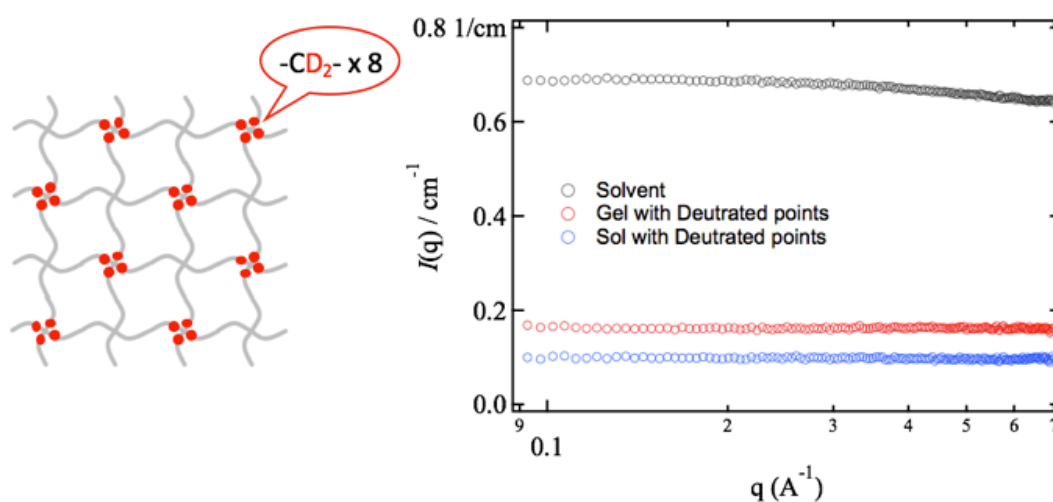


Figure 1. An illustration of tetra-PEG gels, in which deuterium atoms were introduced in the vicinity of the branching point and result of SANS by matching contrast methods.

Renormalization of the spin excitations in multiferroic HoMnO_3 from magnon-phonon coupling

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We carried out inelastic neutron scattering experiments to investigate the magnetic excitation spectrum of multiferroic hexagonal HoMnO_3 . RMnO_3 with rare earths such as $\text{R}=\text{Y}$ or Lu form in a 2D triangular lattice antiferromagnet (TLAF), but with $\text{R}=\text{Ho}$ it is as close as possible to an ideal manifestation of a triangular lattice [1]. Unlike the $\text{R}=\text{Y}$ or Lu cases, a magneto-elastic mode originating from magnon-phonon coupling [2] was not detected in spin wave dispersion of HoMnO_3 . And we couldn't find any noticeable two magnon signal from magnon-magnon interaction. However, it was observed that there is a renormalization of magnon energy levels. We employ linear spin wave theory (LSWT), Heisenberg XXZ model including anharmonic term, and Einstein Site Phonon (ESP) models [3, 4] in order to accurately describe the entire magnon spectra. We are lead to the conclusion that ESP model, which includes magnon-phonon coupling in the Hamiltonian, is the best model to describe magnon dispersion curves as well as the dynamical structure factor. We deduced that the magnon-phonon interactions are still stronger than magnon-magnon interactions in an ideal TLAF.

[1] X. Fabrèges et al, *Phys. Rev. Lett.* **103**, 067204 (2009)

[2] J. Oh et al, *Nat. Commun.* **7**, 13146 (2016)

[3] D. L. Bergman et al, *Phys. Rev. B* **74**, 134409 (2006)

[4] F. Wang and A. Vishwanath, *Phys. Rev. Lett.* **100**, 077201 (2008)

Structural analysis of conversion-controlled critical clusters gel by small angle neutron scattering

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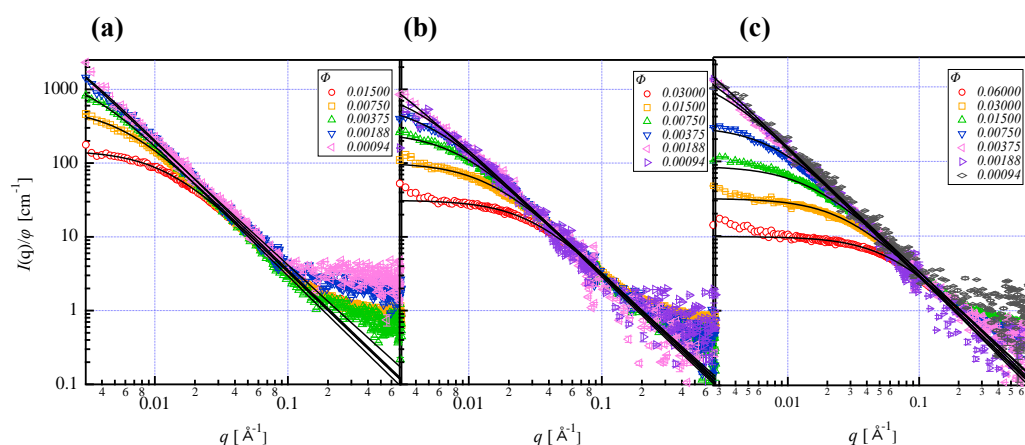
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Polymer gels can be synthesized by various chemical methods. Recently, we established a gelation system by mixing two type of tetra-functional prepolymers, which have complementary reactive end-groups with the other type of prepolymers [1,2]. In this study, we carried out small angle neutron scattering (SANS) study to investigate the critical clusters on way become a gel by quenching the gelation reaction near the gel point. We prepared two kinds of Tetra-PEGs are mixed by the stoichiometric molar ratio in D₂O buffer at the various final concentration of 15, 30, 60 g/L. The reaction bath was quenched at near the gel point by adding small amount of HCl solution into the reaction bath to make pH close to 0, in which the reaction of amine and NHS stops. And then, each resultant critical solution were diluted into different level (60, 30, 15, 7.5, 3.75, 1.88, 0.94 g/L) to fully characterize the structure and the size-distribution of the critical clusters.

[1] K. Hayashi *et al.*, *Nature Biomedical Engineering* 1(3), 0044 (2017).

[2] Li, X. *et al.*, *Macromolecules* 50, 3655 (2017).



The SANS profiles of critical cluster. An illustration of (a) $(r, \phi) = (0.5, 0.015)$ and (b) $(r, \phi) = (0.5, 0.030)$ and (c) $(r, \phi) = (0.5, 0.060)$

Crystal and higher-ordered structure of bi-drawn polyethylene films with neutron and X-ray scattering

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Introduction

For making commercial films, the biaxial stretching processes were widely used. The deformation processes of the polymer chain may be not detailed clearly. In this study, we focused on molecular weight dependence on nm- and micron-scaled morphology with wide-angle, small-angle X-ray and small-angle neutron scattering measurements.

Experiments

Protonated polyethylene and deuterated polyethylene were dissolved into hot o-dichlorobenzene and poured into methanol, therefore, were completed blended. Sample A, B and C are deuterated polyethylene (PE) ($M_w = 600k$), blend PE of protonated ($M_w = 2M$) and deuterated ones of weight ratio at 3:97, and blend PE of protonated ($M_w = 58k$) and deuterated ones at 3:97, respectively. Each sample was drawn simultaneously and sequentially.

Results

Figures 1 and 2 show SAXS and SANS profiles of simultaneously bi-axially drawn films of 2×2. There was hardly any difference between A, B and C. Therefore, we could find independence of molecular weight for morphology of simultaneously bi-axially drawn films. On the other hands, sequentially biaxial stretching, films of 3×1, 3×1.3, 3×1.6, 3×2 were observed. In the presentation, we would like to discuss about difference between simultaneously and sequentially bi-axial drawn films.

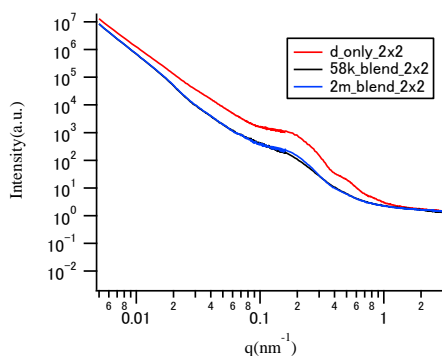


Figure 1. SAXS profiles of simultaneously bi-axial drawn films.

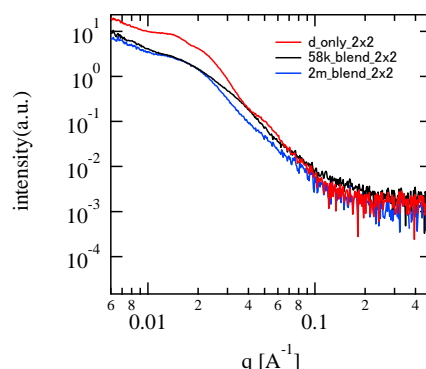


Figure 2. SANS profiles of simultaneously bi-axial drawn films.

Microstructure and Rheology of Graphene Oxide in Polymer Solutions with Varying Molecular Weight

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Graphene oxide (GO) has been attracted great interest in material science, based on its extraordinary mechanical and electrical properties. Compared to graphene, GO is easier to synthesize and has greater approachability and manageability. Another noteworthy aspect of GO aqueous suspension is that it can exhibit liquid crystal (LC) phase, implying potential applications to the electronic and optical devices. While there is a considerable amount of studies on the improved macroscopic property of GO applications using LC phase, the understanding on microscopic behavior of GO in solutions is still lacking.

In this study, we examine the detailed microstructure and rheological property of GO suspensions, focusing on their dispersions in solutions. A remarkable result is that adding poly(ethylene glycol) (PEG) into GO suspensions can improve the stability and reduce the viscosity of suspension. Extensive small angle x-ray scattering and rheology studies were employed to investigate the effect of PEG molecular weight on the structure and LC property of GO suspension.

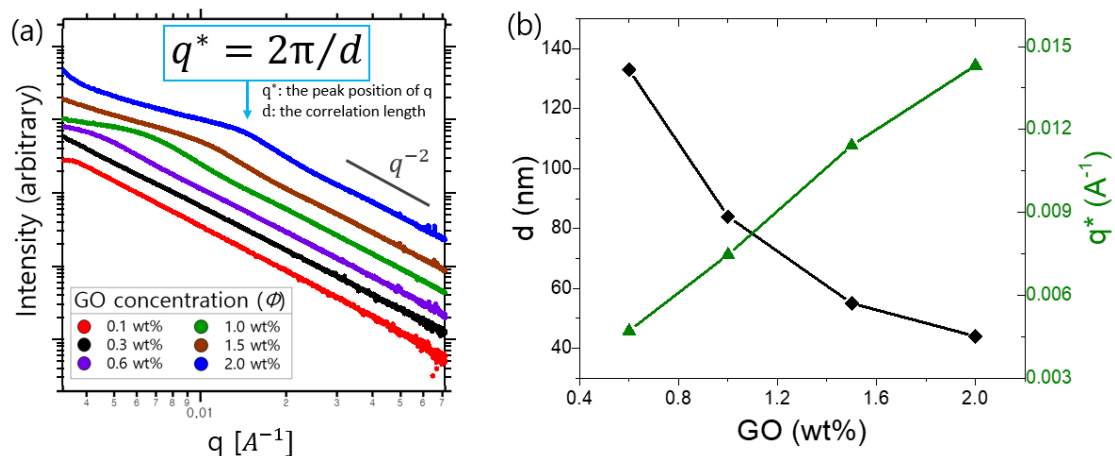


Figure 1. The correlation length with GO concentration. (a) Scattering intensity vs. q -vector and (b) calculated average-correlation length.

Structural analysis of physical gel cross-linked by double-stranded DNA by SANS

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Tetra-PEG gel^[1] is a chemical gel which is synthesized by end-crosslinking two complementary four-armed macromers: Tetra-PEG (polyethylene glycol). It has ideal homogeneous network because the branching points are uniformly distributed, which has been proved by SANS study^[2]. Recently, we succeeded in synthesis of a new type of Tetra-PEG gel which is crosslinked by double-stranded DNA (Figure 1). It is a highly selective physical bonding, thus the gel is expected to have more homogenized polymer network and more controllable crosslinking points than conventional physical gels, such as agarose gel or gelatin gel.

We carried out small angle neutron scattering(SANS) experiment on the new physical gel at various temperatures to investigate the network structure and its structural change along sol-gel transition. Some results are shown in Figure 2. There are some features observed: *a)* At any temperature, the form of double-stranded or single stranded DNA is not observed, because the difference of the scattering length densities of DNA and solvent is much lower than that of PEG polymer and solvent. *b)* When temperature is low enough, the gel has as homogeneous network as normal tetra-PEG gel. *c)* By elevating temperature, the scattering intensity became larger, while no significant change was observed around melting temperature of DNA($T_m \sim 63^\circ\text{C}$).

[1] Sakai, T. *et al.*, *Macromolecules* **2008**, 41(14), 5379

[2] Matsunaga, T. *et al.*, *Macromolecules* **2009**, 42(16), 1344

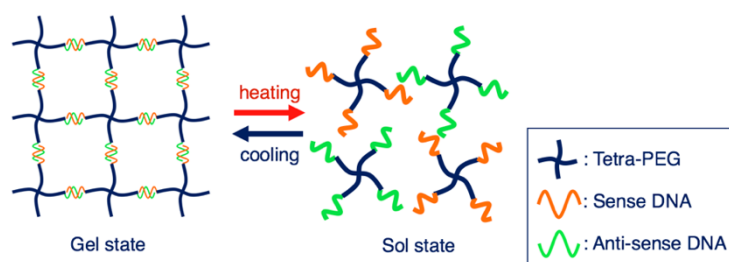


Figure 1. An illustration of sol-gel transition of physical gel crosslinked with double-stranded DNA.

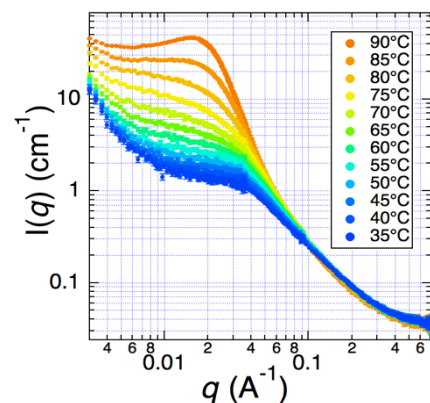


Figure 2. The result of SANS at elevating and lowering temperatures.

Synthesis of Monodisperse Single-Crystalline Gold Nanoparticles

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Polyhedral gold nanoparticles are of great interest in a wide range of applications due to their unique optical and chemical properties, which are induced by well-defined facets, corners, and size. While various polyhedral gold nanoparticles have been mainly synthesized by the growth of single crystalline spheres using a seed-mediated method, synthesis of the single crystalline spheres itself remains challenging due to the complicated synthetic procedures. For examples, the single crystalline spheres have been made by etching of gold nanorods or further growth of octahedrons. Therefore, the development of a simple and easy approach to synthesize the single crystalline spheres is always desirable.

In this study, we present the seedless method of synthesizing monodisperse single-crystalline gold spheres and cuboctahedron [1] using cetyltrimethylammonium 4-vinylbenzoate (CTVB) as a selective capping and a reducing agent in the presence of silver nitrate (AgNO_3) and hydrochloric acid (HCl). This single crystalline gold sphere can be used as a seed for the polyhedral nanoparticles.

[1] A. Umar and S.-M. Choi et al., Chemistry of Materials 28, 4962 (2016)

Microstructural characterization of a Japanese sword using neutron diffraction

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Japanese swords are paid great admiration in the world as the honour of classical technology and of the beautiful art. There have been reports in the past that tried to elucidate this Japanese sword's microstructure using the microscopy analyses and X-ray diffraction (XRD) on the cross sections of swords after mechanical cutting [1]. This observation style was possible in the past, but at the present time when Japanese swords became valuable, non-destructive testing is desired. A mapping measurement using pulsed neutron diffraction (ND) with time-of-flight (TOF) method is suitable for the non-destructive microstructural studies in Japanese swords because of the high penetration ability of neutron and the simultaneous measurement of multiple Bragg peaks. Careful analysis of the Bragg peaks in a neutron diffraction pattern can reveal important structural details of a sample material such as internal stresses, phase conditions, dislocations, texture etc.

The microstructural observation in a Japanese sword made in Keicho era (1596-1615) was performed by an ND mapping using TAKUMI, a TOF neutron diffractometer for engineering materials studies at J-PARC. The appearance of the sword and the measured positions are shown in Fig. 1. The distributions of phase, lattice constant, residual strain, phase fraction, dislocation density and crystallite size were evaluated from the ND patterns. The details will be reported.

[1] For instance: T. Takahashi et al., Discovering New Aspects in a Japanese Sword. *Tetsu-to-Hagane* 71 (1985) 108-114.



Appearance of a Japanese sword and the positions used in the ND mapping

Present Status of University-owned Neutron Beam Instruments at JRR-3

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ISSP owns 14 neutron-beam instruments in the Japan Research Reactor No. 3 (JRR-3), which has been built and owned by Japan Atomic Energy Agency (JAEA), and operates a general user-program for neutron scattering research for half a century. Unfortunately, the operation of JRR-3 has been suspended since the Great East-Japan Earthquake disaster on Mar. 11, 2011. To restore the JRR-3 operation, many tasks of safety management must be met the government regulation. We have been proceeding mainly two following tasks in a recent few years.

One is cleaning up and rearrangement of a radiation-controlled laboratory in JRR-3. Users-owned items were returned to the owners, and less-frequently-used accessories were moved to outside of the laboratory. All the items continuously-preserved in JRR-3 have been registered in a database with the owner, installation location, period and specific character are recorded.

The other is an organization of an inter-facility safety-and-health control team. In order to transfer information from the facility owner, JAEA, to the instrument staff members, we organized “University-owned instruments Safety-management Team (UST)” which is comprised of one professor and technical staff members. Under the leadership of JAEA, the UST have cooperatively proceeded comprehensive safety activities such as check of electric equipment and item management. For our instruments, commissioning has been carried out together with the responsible instrument staff members, toward the upcoming restoration of the reactor operation.

Current status of BL06 VIN ROSE at J-PARC Material Life Science Facility

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The two neutron spin echo spectrometers with resonance neutron spin flippers, that is, a neutron resonance spin echo (NRSE) instrument and a modulated intensity by zero effort (MIEZE) instrument, have been installed by High Energy Accelerator Research Organization (KEK) and Kyoto University at J-PARC Material Life Science Facility occupying BL06. NRSE is suitable to study slow dynamics of soft condensed matter with high energy resolution, while MIEZE has a big advantage of flexible sample environments especially for magnetic field application at the sample position. The designed dynamic range of MIEZE is $0.2 < Q[\text{\AA}^{-1}] < 3.5$ and $0.001 < t[\text{ns}] < 2$, and that of NRSE is $0.02 < Q[\text{\AA}^{-1}] < 0.65$ and $0.1 < t[\text{ns}] < 100$, respectively [1].

In FY2014, the first neutron beam was accepted, and the characteristics of the neutron guides were quantitatively investigated [2]. The first MIEZE signal was observed at the end of FY2014. Since then, various improvements and developments for the public use of BL06 have advanced. At the NRSE spectrometer, the two-dimensional ellipsoidal neutron-focusing supermirrors are now under development in close collaboration with the RIKEN center for advanced photonics.

In the presentation, the current status of the MIEZE and NRSE spectrometers at BL06 will be summarized

[1] M. Hino et al. Physics Procedia 2013, **42**, 136-141.

[2] M. Hino et al. J. Nucl. Sci. Tech. 2017, **54**, 1223-1232.

Local structure observation of Sm doped LaB₆ by white neutron atomic resolution holography

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Physical properties of materials are often controlled by doping. Thus, understanding the local structure such as distorted structures around the dopant is important to clarify the origins of properties; it is also indispensable for development of functional materials. To observe local structures, having no translational symmetry, the best probe is atomic resolution holography (ARH) using x-rays and photoelectrons. ARH can observe local structures in a large three-dimensional area. On the other hand, it is difficult to observe light elements such as H, B or O. Therefore, we have developed

ARH using white neutrons at J-PARC, which are sensitive to light elements. We are trying to apply this novel technique to various fields of materials science. In this study, we focus on a strongly correlated electron system RB₆ (R: rare earth). When Sm is doped in LaB₆ and YbB₆, it behaves as Sm²⁺ in the former and as Sm³⁺ in the latter [1]. We expect that the difference of local structures around Sm may play an important role in this change of valency. White neutron holography experiments were performed on a single crystal La¹¹B₆ with 2% doped Sm. Fig. 1 is an atomic image of the (100) plane around Sm, obtained by neutron ARH. The atomic images were reconstructed using data in the neutron energy range from 10 meV to 200 meV. As shown in Fig.1, La around the Sm dopant are located at the same positions as in pure LaB₆. Moreover, we also succeeded in reconstructing atomic image of ¹¹B (no figure). These results indicate that neutron ARH is an effective probe for investigations of local structures even for Sm doped rare earth borides, which are attracted much interest in physics of magnetism.

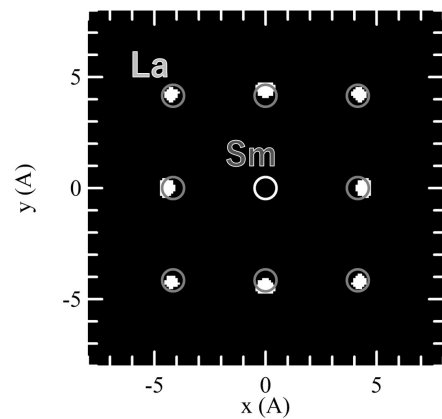


Fig. 1 Atomic image of
La around Sm in Sm doped

[1] J.M. Trascon, et al., J. Physique, 41 (1980) 1135.

Status of the high intensity total diffractometer, NOVA, at J-PARC.

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A neutron total diffractometer, NOVA, was constructed at J-PARC (Japan Proton Accelerator Research Complex) to analyze crystalline and non-crystalline structure of hydrogen storage materials in the Q range from 0.03 \AA^{-1} to 100 \AA^{-1} . By adapting neutron total scattering technique to hydrogen storage materials, it is aimed to extract of variety of structural information such as hydrogen-hydrogen correlation in materials.

It is feasible to obtain atomic pair distribution function which derived by the Fourier transformation of observed static structure factor, $S(Q)$, is reliable up to 200 \AA . It was also shown that NOVA is a powerful instrument to observe structure of materials under in-situ conditions. In-situ sample environments such as Temperature controlled sample changer (no. of samples per load is 18 and temp. range is $20 \sim 750 \text{ K}$), H_2/D_2 gas atmosphere (Max gas pressure is 10 MPa , temperature range is $50 \text{ K} \sim 473 \text{ K}$), low-temperature ($\sim 2 \text{ K}$) and high-temperature ($\sim 1373 \text{ K}$) and so on are available on NOVA. On-line data reduction scheme has been equipped and user interface is now developing. Variety of materials like hydrogen storage materials, magnetic materials, super conducting materials, liquid and glassy materials were investigated. Recent results of structure analysis on NOVA will be presented.

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Investigations of Local Structures by White Neutron Atomic Resolution Holography with γ -ray Energy Analysis

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For functional materials, the properties are controlled by impurity doping. In such materials, the change of local structures around the dopants plays an important role. Atomic resolution holography (ARH) with x-rays and electrons is effective to observe local structures, which has been already established in materials science. We are developing white neutron ARH at J-PARC to investigate local atomic structures around light elements. In the works, to obtain pure signals, we performed ARH experiments using a Ge detector, which has a high energy resolution. Fig.1 shows results of atomic images around B in 0.26 at% ^{10}B doped Si in the (100) plane (upper panel), and (100) plane of the nearest neighbor position (lower panel). Z is the distance to the planes from the doped B. Atomic images are observed exactly at the P1 and P2 positions, while atomic images at P3 positions are shifted. The reasons of the shifts are now under considerations.

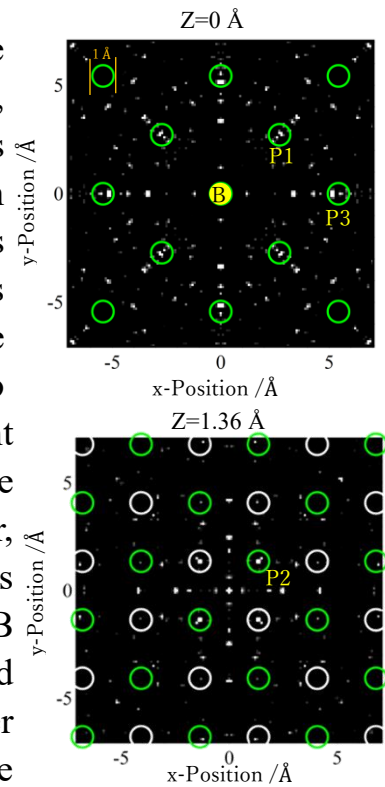


Fig.1 Atomic image around B in 0.26 at% B doped Si. The circles are expected positions of Si.

Local structure investigations of B doped Si by neutron atomic-resolution holography

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Some functional materials show excellent properties by doping a small amount of foreign elements. A typical example is B doped Si. In such materials, slight changes of the structures around dopants (local structure) are important for the functions. Atomic resolution holography (ARH) can observe the local structures within 20 Å from the selected atom (dopant) in a three dimensional area. ARH has already been established using x-rays and photoelectrons. On the contrary, ARH reproduces artifacts in principle [1]. Note that such artifacts can be avoided by using multi wavelengths. We are developing ARH using white neutrons at J-PARC, which can obtain the holograms of 130 different wavelengths at once using the time of flight technique. In the present study, we measured the atomic structures around B in single crystal Si with 0.13 at% ¹⁰B. Fig. 1 is the atomic image in the (100) plane centering doped B. The other circles are the expected positions of Si. As shown in Fig. 1, the atomic images of Si are observed at the nearest expected positions in the plane, indicating that most of the doped B are located at the substitutional site. Our poster will report the principles of holography, and the latest experimental results.

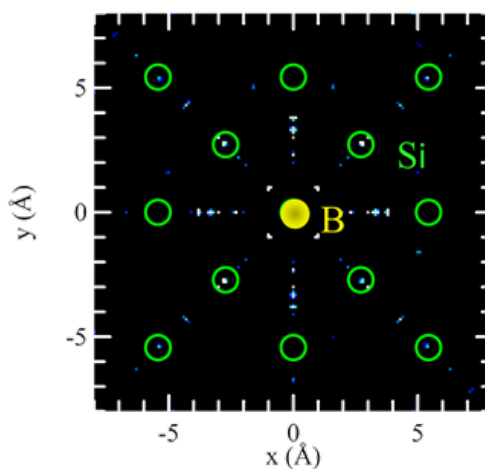


Fig. 1 The atomic image in the (100) plane around B. The circles indicate expected Si positions.

[1] K. Hayashi et al., J. Phys.: Condens. Matter, **24** (2012) 9320.

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