Publications

Division of New Materials Science

Takigawa group

We have been performing nuclear magnetic resonance experiments on various quantum spin systems and strongly correlated electron systems to explore novel quantum phases with exotic ordering and fluctuation phenomena. The major achievements in the year 2014 include: (1) Investigation by 11B-NMR of the real space imaging of a spin polaron generated by non-magnetic defects in the Shastry-Sutherland spin system $SrCu_2(BO_3)_2$, (2) Microscopic examination by angle resolved 27Al-NMR of the quarupole order and mutipolar fluctuations in $PrTi_2Al_{20}$, (3) Continued investigation on single crystals of volborthite, a frustrated quantum spin system with distorted Kagome lattice aimed at the full understanding of the phase diagram and magnetic structure in magnetic field.

1. †*Field-induced incommensurate phase in the strong-rung spin ladder with ferromagnetic legs: H. Yamaguchi, H. Miyagai, M. Yoshida, M. Takigawa, K. Iwase, T. Ono, N. Kase, K. Araki, S. Kittaka, T. Sakakibara, T. Shimokawa, T. Okubo, K. Okunishi, A. Matsuo and Y. Hosokoshi, Phys. Rev. B **89** (2014) 220402(R)(1-7).

2. †Search for a spin-nematic phase in the quasi-one-dimensional frustrated magnet $LiCuVO_4$: N. Büttgen, K. Nawa, T. Fujita, M. Hagiwara, P. Kuhns, A. Prokofiev, A. P. Reyes, L. E. Svistov, K. Yoshimura and M. Takigawa, Phys. Rev. B **90** (2014) 134401(1-7).

3. *Novel Phase Transitions in the Breathing Pyrochlore Lattice:^{7}Li-NMR on $LiInCr_4O_8$ and $LiGaCr_4O_8$: Y. Tanaka, M. Yoshida, M. Takigawa, Y. Okamoto and Z. Hiroi, Phys. Rev. Lett. **113** (2014) 227204(1-5).

4. †Real Space Imaging of Spin Polarons in Zn-Doped $SrCu_2(BO_3)_2$: M. Yoshida, H. Kobayashi, I. Yamauchi, M. Takigawa, S. Capponi, D. Poilblanc, F. Mila, K. Kudo, Y. Koike and N. Kobayashi, Phys. Rev. Lett. **114** (2015) 056402(1-5).

Sakakibara group

We study magnetism and superconductivity of materials having low characteristic temperatures. These include heavy-electron systems, quantum spin systems and frustrated spin systems. The followings are some selected achievements in the fiscal year 2014. (1) Field and temperature variations of the specific heat $C(H,T)$ of the heavy fermion superconductor UBe_{13} (T_c=0.9 K) were examined at temperatures down to 80 mK. Quite unexpectedly, the magnetic-field dependence of $C(H)$ is linear in H with no angular dependence at low fields in the superconducting state, implying that the gap is fully open over the Fermi surfaces, in stark contrast to the previous expectation. (2) Magnetization and magnetic torque of a high-quality single crystal of Sr_2RuO_4 have been measured down to 100 mK under a precise control of the magnetic-filed orientation. When the magnetic field is applied exactly parallel to the ab plane, a sharp magnetization jump at the upper critical field is observed at low temperatures, evidencing the first-order superconducting-normal transition. A strong magnetic torque appearing when H is slightly tilted away from the ab plane confirms the intrinsic anisotropy $\Gamma = \xi_a / \xi_c$ of as large as 60 even at 100 mK. The present results raises fundamental issues in both the existing spin-triplet and spin-singlet scenarios.

1. †Fine-Tuning of Magnetic Interactions in Organic Spin Ladders: H. Yama80guchi, H. Miyagai, T. Shimokawa, K. Iwase, T. Ono, Y. Kono, N. Kase, K. Araki, S. Kittaka, T. Sakakibara, T. Kawakami, K. Okunishi and Y. Hosokoshi, J. Phys. Soc. Jpn. **83** (2014) 033707(1-4).

1. Novel Electronic States of Heavy Fermion Compound $YbCo_2Zn_{20}$: F. Honda, Y. Taga, Y. Hirose, S. Yoshiuchi, Y. Tomooka, M. Ohya, J. Sakaguchi, T. Takeuchi, R. Settai, Y. Shimura, T. Sakakibara, I. Sheikin, T. Tanaka, Y. Kubo and Y. Onuki, J. Phys. Soc. Jpn. **83** (2014) 044703(1-9).

2. †Possible Evolution of Antiferromagnetism in Zn-Doped Heavy-Fermion Superconductor $CeCoIn_5$: M. Yokoyama, K. Fujimura, S. Ishikawa, M. Kimura, T. Hasegawa, I. Kawasaki, K. Tenya, Y. Kono and T. Sakakibara, J. Phys. Soc. Jpn. **83** (2014) 033706(1-5).

3. Thermodynamic Study of Nodal Structure and Multiband Superconductivity of KFe_2As_2: S. Kittaka, Y. Aoki, N. Kase, T. Sakakibara, T. Saito, H. Fukazawa, Y. Kohori, K. Kihou, C. Ho Lee, A. Iyo, H. Eisaki, K. Deguchi, N. K. Sato, Y.

* Joint research among groups within ISSP.

Tsutsumi and K. Machida, J. Phys. Soc. Jpn. **83** (2014) 013704(1-4).

4. †*Field-induced incommensurate phase in the strong-rung spin ladder with ferromagnetic legs: H. Yamaguchi, H. Miyagai, M. Yoshida, M. Takigawa, K. Iwase, T. Ono, N. Kase, K. Araki, S. Kittaka, T. Sakakibara, T. Shimokawa, T. Okubo, K. Okunishi, A. Matsuo and Y. Hosokoshi, Phys. Rev. B **89** (2014) 220402(R)(1-7).

5. Sharp magnetization jump at the first-order superconducting transition in Sr_2RuO_4: S. Kittaka, A. Kasahara, T. Sakakibara, D. Shibata, S. Yonezawa, Y. Maeno, K. Tenya and K. Machida, Phys. Rev. B **90** (2014) 220502(R) (1-5).

6. Measurement of the spin-orbit coupling in superconducting Sr_2RuO_4 using polarized light and spin-resolved photoemission spectroscopy: Evidence for a breakdown in the singlets and triplets pairing mechanisms: C. N. Veenstra, Z. -H. Zhu, M. Raichle, B. M. Ludbrook, A. Nicolaou, B. Slomski, G. Landolt, S. Kittaka, Y. Maeno, J. H. Dil, I. S. Elfimov, M. W. Haverkort and A. Damascelli, Phys. Rev. Lett. **112** (2014) 127002(1-4).

7. Multiband superconductivity with unexpected deficiency of nodal quasiparticles in $CeCu_2Si_2$: S. Kittaka, Y. Aoki, Y. Shimura, T. Sakakibara, S. Seiro, C. Geibel, F. Steglich, H. Ikeda and K. Machida, Phys. Rev. Lett. **112** (2014) 067002(1-5).

8. Spin-Chirality-Driven Ferroelectricity on a Perfect Triangular Lattice Antiferromagnet: H. Mitamura, R. Watanuki, K. Kaneko, N. Onozaki, Y. Amou, S. Kittaka, R. Kobayashi, Y. Shimura, I. Yamamoto, K. Suzuki, S. Chi and T. Sakakibara, Phys. Rev. Lett. **113** (2014) 147202 (1-5).

9. Anisotropic Superconductivity of the Caged Compound $Y_5Rh_6Sn_{18}$ with Unusual Normal-State Electrical Resistivity: N. Kase, S. Kittaka, T. Sakakibara and J. Akimitsu, JPS Conf. Proc. **3** (2014) 015042 (1-5).

10. †*Low Temperature Magnetic Properties of Frustrated Quantum Spin Chain System $Rb_2Cu_2Mo_3O_{12}$: Y. Yasui, R. Okazaki, I. Terasaki, M. Hase, M. Hagihala, T. Masuda and T. Sakakibara, JPS Conf. Proc. **3** (2014) 014014 (1-6).

11. Low Temperature Magnetization of Yb_2Pt_2Pb Along the Hard Magnetization Axis: Y. Shimura, T. Sakakibara, K. Iwakawa, K. Sugiyama and Y. Onuki, JPS Conf. Proc. **3** (2014) 014029 (1-6).

12. *Magnetization and Specific Heat of the Caged Compound PrV_2Al_{20}: K. Araki, Y. Shimura, N. Kase, T. Sakakibara, A. Sakai and S. Nakatsuji, JPS Conf. Proc. **3** (2014) 011093(1-5).

13. Magnetization Study of the Quantum Critical Behavior of the One Dimensional Spin-1/2 Heisenberg Antiferromagnet CuPzN: Y. Kono, T. Sakakibara, C. Aoyama, M. M. Turnbull, C. Landee and Y. Takano, JPS Conf. Proc. **3** (2014) 012015 (1-5).

14. †Metal-Insulator Transition in Pyrochlore Oxide $(Nd_{1-x}Pr_x)_2Ir_2O_7$ $(0.7 \leq x \leq 1)$: K. Matsuhira, K. Kuroda, T. Sakakibara, M. Wakeshima and Y. Hinatsu, JPS Conf. Proc. **3** (2014) 013017 (1-6).

15. †Quantum phase near the saturation field in the S=1/2 frustrated spin ladder: H. Yamaguchi, H. Miyagai, Y. Kono, S. Kittaka, T. Sakakibara, K. Iwase, T. Ono, T. Shimokawa and Y. Hosokoshi, Phys. Rev. B **91** (2015) 125104 (1-5).

16. †S=1/2 ferromagnetic-antiferromagnetic alternating Heisenberg chain in a zinc-verdazyl complex: H. Yamaguchi, Y. Shinpuku, T. Shimokawa, K. Iwase, T. Ono, Y. Kono, S. Kittaka, T. Sakakibara and Y. Hosokoshi, Phys. Rev. B **91** (2015) 085117 (1-6).

17. Field-Induced Quantum Criticality and Universal Temperature Dependence of the Magnetization of a Spin-1/2 Heisenberg Chain: Y. Kono, T. Sakakibara, C. P. Aoyama, C. Hotta, M. M. Turnbull, C. P. Landee and Y. Takano, Phys. Rev. Lett. **114** (2015) 037202 (1-5).

18. Field-Orientation Dependence of Low-Energy Quasiparticle Excitations in the Heavy-Electron Superconductor UBe_{13}: Y. Shimizu, S. Kittaka, T. Sakakibara, Y. Haga, E. Yamamoto, H. Amitsuka, Y. Tsutsumi and K. Machida, Phys. Rev. Lett. **114** (2015) 147002 (1-5).

19. *Antiferromagnetic transition of the caged compound $TmTi_2Al_{20}$: N. Kase, Y. Shimura, S. Kittaka, T. Sakakibara, S. Nakatsuji, T. Nakano, N. Takeda and J. Akimitsu, J. Phys.: Conf. Ser. **592** (2015) 012052.

Mori group

We have successfully developed and characterized the functional molecular materials. The major achievements in 2014 are (1) to discover the conductivity and magnetism switching caused by coupled deuterium and electron transfer in the organic single-component crystal κ-D_3(Cat-EDT-TTF)$_2$, (2) to find the solid-solid interconversion with dynamic change of donor arrangement and electrical properties related to bending of hydrogen-bonded unit for the organic crystal β'-H_3(Cat-EDO-TTF)$_2BF_4$, and (3)to observe the anisotropic transport behavior under uniaxial strain in the pressure indeuced superconductor

† Joint research with outside partners.

β-(*meso*-DMBEDT-TTF)$_2$PF$_6$. The introduction of a large variety of molecule degree of freedom to solid promises the development of new trends in functional molecular materials.

1. Gapless Quantum Spin Liquid in an Organic Spin-1/2 Triangular-Lattice κ-H$_3$(Cat-EDT-TTF)$_2$: T. Isono, H. Kamo, A. Ueda, K. Takahashi, M. Kimata, H. Tajima, S. Tsuchiya, T. Terashima, S. Uji and H. Mori, Phys. Rev. Lett. **112** (2014) 177201.

2. A biferrocenium salt containing paramagnetic tetracyanoquinodimethane hexamers: charge disproportionation via donor–acceptor interactions: T. Mochida, Y. Funasako, K. Takahashi, M. Inokuchi, T. Sakurai, S. Ikeda, H. Ohta, H. Mori and M. Uruichi, Chem. Commun. **50** (2014) 13370.

3. Solid-solid phase interconversion in an organic conductor crystal: hydrogen-bond-mediated dynamic changes in π-stacked molecular arrangement and physical properties: J. Yoshida, A. Ueda, A. Nakao, R. Kumai, H. Nakao, Y. Murakami and H. Mori, Chem. Commun. **50** (2014) 15557.

4. Hydrogen-Bond-Dynamics-Based Switching of Conductivity and Magnetism: A Phase Transition Caused by Deuterium and Electron Transfer in a Hydrogen-Bonded Purely Organic Conductor Crystal: A. Ueda, S. Yamada, T. Isono, H. Kamo, A. Nakao, R. Kumai, H. Nakao, Y. Murakami, K. Yamamoto, Y. Nishio and H. Mori, J. Am. Chem. Soc. **136** (2014) 12184.

5. Protonation of Pyridyl-Substituted TTF Derivatives: Substituent Effects in Solution and in the Proton-Electron Correlated Charge-Transfer Complexes: S. C. Lee, A. Ueda, A. Nakao, R. Kumai, H. Nakao, Y. Murakami and H. Mori, Chem. Eur. J. **20** (2014) 1909.

6. Synergistic Spin Transition between Spin Crossover and Spin-Peierls-like Singlet Formation in the Halogen-Bonded Molecular Hybrid System: [Fe(Iqsal)$_2$][Ni(dmit)$_2$]·CH$_3$CN·H$_2$O: K. Fukuroi, K. Takahashi, T. Mochida, T. Sakurai, H. Ohta, T. Yamamoto, Y. Einaga and H. Mori, Angew. Chem. Int. Ed. **53** (2014) 1983.

7. Uniaxial Strain Effect of the Moderately Dimerized Molecular Conductor β-(*meso*-DMBEDT-TTF)$_2$PF$_6$: Y. Nishida, T. Isono, A. Ueda and H. Mori, Eur. J. Inorg. Chem. **2014** (2014) 3845.

8. Dynamics of Charge Ordering in the Nonlinear Regime of θ-(BEDT-TTF)$_2$CsZn(SCN)$_4$: M. Abdel-Jawad, I. Terasaki, T. Mori and H. Mori, J. Phys. Soc. Jpn. **84** (2015) 033707.

9. Phase-change memory function of correlated electrons in organic conductors: H. Oike, F. Kagawa, N. Ogawa, A. Ueda, H. Mori, M. Kawasaki and Y. Tokura, Phys. Rev. B **91** (2015) 041101.

10. 電子とプロトンが協奏する新しい分子機能性結晶の展開 : 森 初果 , 上田 顕 , 磯野 貴之 , 固体物理 **49** (2014) 149-160.

11. 化学の力を生かした新しい分子性機能材料の開発 : 森 初果 , 化学 **69**、**No1** (2014) 20-21.

12. 金属状態を示す純有機単成分導体 : 森 初果 , 工業材料 **62**　**No1** (2014) 26-27.

13. Women in Physics に参加して : 森 初果 , 東京大学理学部ニュース　理学エッセイ **No13** (2014) https://www.s.u-tokyo.ac.jp/ja/s.

Nakatsuji group

Our group explores novel quantum phases and phase transitions in rare-earth and transition metal based compounds. The followings are some relevant results obtained in 2014. (1) We have succeeded in growing high quality single crystals of PrV_2Al_{20} and discovered heavy fermion superconductivity due to orbital fluctuations in the antiferro-quadrupolar state. Our results suggest a proximity to the quadrupolar quantum criticality due to the competition between quadrupolar Kondo effect and multipolar ordering. Indeed, we found that the application of the magnetic field suppresses the quadrupolar transition and induces a novel type of quantum criticality. Our results indicate that $PrTr_2Al_{20}$ provides a model system to investigate the quantum criticality purely associated with the multipolar (orbital) degrees of freedom. (2) Our high resolution thermodynamic measurements at very low temperatures on β-$YbAlB_4$ has revealed that two components associated with quantum criticality and heavy fermion behaviour exist in this intermediate valence material. The similar type of heavy fermion component dominates in the properties of α-analog. Future comparative studies will clarify the origin of the spontaneous quantum criticality of the β-phase. (3) Finally, the magnetic domain control of antiferromagnetism is an interesting subject for both basic science and their technological application. We found that the magnetic domains in the well-known antiferromagnetic Mott insulator NiS_2 may be controlled by the cooling procedure under magnetic fields. Further investigation such as the magnetoresistance and Hall resistivity would be important to find out useful functions in these non-collinear antiferromagnets.

1. Electronic Structure of Quantum Spin-liquid Coupound $Ba_3CuSb_2O_9$: T. Sugimoto, T. Mizokawa, H. Wadati, K. Takubo, A. Damascelli, T. Z. Regier, G. A. Sawatzky, N. Katayama, H. Sawa, K. Kimura and S. Nakatsuji, J. Phys. Soc. Jpn. **3** (2014) 014007 (1-4).

---

* Joint research among groups within ISSP.

2. †*Heavy fermion superconductivity under pressure in the quadrupole system $PrTi_2Al_{20}$: K. Matsubayashi, T. Tanaka, J. Suzuki, A. Sakai, S. Nakatsuji, K. Kitagawa, Y. Kubo and Y. Uwatoko, J. Phys. Soc. Jpn. **3** (2014) 011077 (1-5).

3. Sample Dependence of the Quadrupolar Transition in the Nonmagnetic cubic Γ_3 Compound PrV_2Al_{20}: M. Tsujimoto, A. Sakai and S. Nakatsuji, J. Phys. Soc. Jpn. **3** (2014) 011066(1-6).

4. Structural and Magnetic Properties of α-$Yb(Al_{1-x}Fe_x)B_4$ under Hydtostatic Pressure: Y. Sakaguchi, S. Ikeda, K. Kuga, S. Nakatsuji, N. Hirao, Y. Ohishi and H. Kobayashi, J. Phys. Soc. Jpn. **3** (2014) 011059 (1-6).

5. Superconducting properties of the ferroquadrupolar cubic Γ_3 compound $PrTi_2Al_{20}$: A. Sakai, K. Kuga and S. Nakatsuji, J. Phys. Soc. Jpn. **3** (2014) 011066 (1-6).

6. Suppression of the Heavy Fermion State in Magnetic Fields in the Mixed Valent α-$YbAlB_4$: Y. Matsumoto, K. Kentaro and S. Nakatsuji, J. Phys. Soc. Jpn. **3** (2014) 011076(1-5).

7. Two magnetic phases in α-YbAl1-xFexB_4: K. Kuga, S. Suzuki and S. Nakatsuji, J. Phys. Soc. Jpn. **3** (2014) 012013(1-4).

8. $CeCu_2Ge_2$: Challenging our understanding of quantum criticality: B. Zeng, Q. R. Zhang, D. Rhodes, Y. Shimura, D. Watanabe, R. E. Baumbach, P. Schlottmann, T. Ebihara and L. Balicas, Phys. Rev. B **90** (2014) 155101.

9. Experimental realization of a quantum breathing pyrochlore antiferromagnet: K. Kimura, S. Nakatsuji and T. Kimura, Phys. Rev. B **90** (2014) 060414.

10. Heavy-Fermion Superconductivity in the Quadrupole Ordered State of PrV_2Al_{20}: M. Tsujimoto, Y. Matsumoto, T. Tomita, A. Sakai and S. Nakatsuji, Phys. Rev. Lett. **113** (2014) 267001 (1-5).

11. 銅酸化物磁性体におけるスピン・軌道の特異な量子状態 : 澤 博 , 中辻 知 , 固体物理 **49** (2014) 533-543.

12. Quantum criticality in a metallic spin liquid: Y. Tokiwa, J. J. Ishikawa, S. Nakatsuji and P. Gegenwart, Nature Mater. **13** (2014) 356-359.

13. X-ray Photoemission and X-ray Absorption Spectroscopy of Hexagonal $Ba_3CuSb_2O_9$: T. Sugimoto, T. Mizokawa, H. Wadati, K. Takubo, A. Damascelli, T. Z. Regier, G. A. Sawatzky, N. Katayama, H. Sawa, K. Kimura and S. Nakatsuji, J. Phys. Soc. Jpn **63** (2014) 549-550.

14. Magnetic and Thermal Properties of the Single Crystalline $Pr_2Zr_2O_7$ in a [111] field: K. Kimura and S. Nakatsuji, JPS Conf. Proc. **3** (2014) 014027[5 pages].

15. *Magnetization and Specific Heat of the Caged Compound PrV_2Al_{20}: K. Araki, Y. Shimura, N. Kase, T. Sakakibara, A. Sakai and S. Nakatsuji, JPS Conf. Proc. **3** (2014) 011093(1-5).

16. *Magnetization of Yb-based mixed-valent compounds at megagauss fields: T. Terashima, Y. H. Matsuda, K. Kuga, Y. Matsumoto and S. Nakatsuji, J. Phys. Conf. Proc. **3** (2014) 011027(1-5).

17. Conduction electron spin resonance in the α-$Yb_{1-x}Fe_xAlB_4$ ($0 \le x \le 0.50$) and α-$LuAlB_4$ compounds: L. Holanda, G. Lesseux, E. Magnavita, R. Ribeiro, S. Nakatsuji, K. Kuga, Z. Fisk, S. Oseroff, R. Urbano, C. Rettori and P. Pagliuso, J. Phys.: Condens. Matter **27** (2015) 255601/1-5.

18. Field Evolution of Quantum Critical and Heavy Fermi-Liquid Components in the Magnetization of the Mixed Valence Compound β-$YbAlB_4$: Y. Matsumoto, K. Kuga, Y. Karaki, Y. Shimura, T. Sakakibara, M. Tokunaga, K. Kindo and S. Nakatsuji, J. Phys. Soc. Jpn. **84** (2015) 024710(1-7).

19. Magnetization Anomaly due to the Non-Coplanar Spin Structure in NiS_2: T. Higo and S. Nakatsuji, J. Phys. Soc. Jpn. **84** (2015) 053702(1-5).

20. Field-induced quadrupolar quantum criticality in PrV_2Al_{20}: Y. Shimura, M. Tsujimoto, B. Zeng, L. Balicas, A. Sakai and S. Nakatsuji, Phys. Rev. B **91** (2015) 241102/1-5.

21. 電子軌道の量子ゆらぎによる新しい超伝導 : 松本 洋介 , パリティ **30-7** (2015) (2-6).

22. Anisotropic transverse magnetoresistivity in α-$YbAlB_4$: Y. Matsumoto, J. Hong, K. Kuga and S. Nakatsuji, J. Phys.: Conf. Ser. **592** (2015) 012086.

23. Anomalous Enhancement of Seebeck Coefficient in Pr-Based 1-2-20 System with Non-Kramers Doublet Ground States: Y. Machida, T. Yoshida, T. Ikeura, K. Izawa, A. Nakama, R. Higashinaka, Y. Aoki, H. Sato, A. Sakai, S. Nakatsuji, N. Nagasawa, K. Matsumoto, T. Onimaru and T. Takabatake, J. Phys.: Conf. Ser. **592** (2015) 012025.

† Joint research with outside partners.

24. Anomalous specific heat behaviour in the quadrupolar Kondo system PrV_2Al_{20}: M. Tsujimoto, Y. Matsumoto and S. Nakatsuji, J. Phys.: Conf. Ser. **592** (2015) 012023.

25. *Antiferromagnetic transition of the caged compound $TmTi_2Al_{20}$: N. Kase, Y. Shimura, S. Kittaka, T. Sakakibara, S. Nakatsuji, T. Nakano, N. Takeda and J. Akimitsu, J. Phys.: Conf. Ser. **592** (2015) 012052.

26. *High Pressure Measurements of the Resistivity of β -$YbAlB_4$: T. Tomita, K. Kuga, Y. Uwatoko and S. Nakatsuji, J. Phys.: Conf. Ser. **592** (2015) 012019.

27. Shubnikov-de Haas Oscillation in the cubic Γ_3-based heavy fermion superconductor PrV_2Al_{20}: Y. Shimura, M. Tsujimoto, A. Sakai, B. Zeng, L. Balicas and S. Nakatsuji, J. Phys.: Conf. Ser. **592** (2015) 012026.

28. †*Synchrotron X-ray spectroscopy study on the valence state and magnetization in α-$YbAl_{1-x}Fe_xB_4$ (x = 0.115) at low temperatures and high magnetic fields: T. Terashima, Y. H. Matsuda, K. Kuga, S. Suzuki, Y. Matsumoto, S. Nakatsuji, A. Kondo, K. Kindo, N. Kawamura, M. Mizumaki and T. Inami, J. Phys.: Conf. Ser. **592** (2015) 012020 (6 pages).

29. High-Field Multi-Frequency ESR in the Rare-Earth Spinel Compound CdYb2S4: D. Yoshizawa, T. Kida, S. Nakatsuji, K. Iritani, M. Halim, T. Takeuchi and M. Hagiwara, Appl. Magn. Reson. **NIL** (2015) 0937-9347.

30. Absence of Jahn-Teller transition in the hexagonal $Ba_3CuSb_2O_9$ single crystal: N. Katayama, K. Kimura, Y. Han, J. Nasu, N. Drichko, Y. Nakanishi, M. Halim, Y. Ishiguro, R. Satake, E. Nishibori, M. Yoshizawa, T. Nakano, Y. Nozue, Y. Wakabayashi, S. Ishihara, M. Hagiwara, H. Sawa and S. Nakatsuji, Proceedings of the National Academy of Sciences (2015), in print.

Division of Condensed Matter Theory

K. Ueda group

Metal-insulator (MI) transition is one of the most intriguing phenomena of the strongly correlated electron systems. Nature of the MI transitions depends on lattice structure even if the orbital degeneracy is not taken into account. We investigate the MI transitions of the Hubbard model on the one-fifth-depleted square lattice by using the cellular dynamical mean field theory, assuming paramagnetic phases. It is shown that the MI transition is of the first order on the plaquette side of the parameter space, while it is continuous on the dimer side. The continuous MI transition is a Lifshitz transition driven by the electron-electron interaction.

1. Continuous Mott transition in a two-dimensional Hubbard model: Y. Yanagi and K. Ueda, Phys. Rev. B **90** (2014) 085113.

2. Magnetic Phases and Edge States of the 1/5-Depleted Square-Lattice Hubbard Model at 1/4 Filling: Y. Yamashita, M. Tomura, Y. Yanagi and K. Ueda, JPS Conf. Proc **3** (2014) 016012.

3. Quantum Phase Transition in the 1/5 Depleted Square Lattice Hubbard Model: Y. Yanagi and K. Ueda, JPS Conf. Proc **3** (2014) 013005.

Takada group

Employing several techniques including the Green's-function approach, quantum Monte Carlo simulations, and several types of variational approaches, we are studying various aspects of quantum many-body problems in condensed matter physics, based mainly on the first-principles Hamiltonian. This year we have obtained a couple of outstanding results: (1) We have revealed that the system of a proton embedded in the electron gas with the density parameter r_s in the region between 2 and 12.5 yields a Kondo spin-singlet resonance state with the Kondo temperature T_K well beyond 1,000K as its ground state, indicating that if a macroscopic number of protons are embedded in a regular array into a metal in this density region to form a periodic Kondo lattice, superconductivity is expected to occur at a temperature as high as $T_K/10$ by referring to heavy-fermion physics. (2) In the electron gas with the density as low as r_s~20, a new phase transition is found to occur as signaled by the anomalous behavior in the momentum distribution function, along with the unusual property of vanishing effective mass at the Fermi level m*~0, leading to a new filed of “light-fermion physics”.

1. Structural evolution of the one-dimensional spectral function from the low- to the high-energy limit: H. Maebashi and Y. Takada, Phys. Rev. B **89** (2014) 201109(R) (1-5).

2. Generic Features of an Electron Injected into the Luttinger Liquid: H. Maebashi and Y. Takada, J Supercond Nov Magn

* Joint research among groups within ISSP.

28 (2015) 1331-1335.

3. 第1原理からの超伝導理論：高田 康民，物性研究 電子版 **Vol.3, No.1** (2014) 031203(1-29).

4. Theory for Reliable First-Principles Prediction of the Superconducting Transition Temperature: Y. Takada, in: *Carbon-based New Superconductors: Toward high-Tc superconductivity (ISBN 978-981-4303-30-9 (Hardcover), 978-981-4303-31-6 (eBook))*, Ch 8, edited by J. Haruyama, (Pan Stanford Publishing Pte. Ltd., 2015), pp. 193-230.

5. Theory of Superconductivity in Graphite Intercalation Compounds: Y. Takada, in: *Materials Science and Materials Engineering*, edited by S. Mahfoudh and M. Nicholls, (Elsevier, 2015), in print.

Oshikawa group

We studied a wide range of fundamental problems in condensed matter theory and statistical mechanics. In particular, we studied the total orbital angular momentum of chiral superfluids. Superfluids/superconductors with vanishing viscosity or resistivity often arise as a result of condensation of pairs of the constituent fermions. Usually, each pair does not involve a circular motion and has zero angular momentum. However, in the superfluid-A phase of liquid Helium-3, for example, each pair is rotating in a particular orientation around its center. We may then expect that the superfluid carries a total angular momentum, i.e. is rotating as a whole. There have been, however, conflicting arguments, and the problem has remained unresolved for more than 30 years. We studied this long-standing question carefully, to find a rather surprising answer: the superfluid indeed has a large total angular momentum, only if each pair has the minimal angular momentum 1. When each pair carries a higher angular momentum (2, 3, 4, ...), the total angular momentum almost vanishes. This counter-intuitive phenomenon is a consequence of subtle quantum mechanical nature of the superfluids.

1. †Quantum criticality in an asymmetric three-leg spin tube: A strong rung-coupling perspective: Y. Fuji, S. Nishimoto, H. Nakada and M. Oshikawa, Phys. Rev. B **89** (2014) 054425 (1-13).

2. †Valence bond distribution and correlation in bipartite Heisenberg antiferromagnets: D. Schwandt, F. Alet and M. Oshikawa, Phys. Rev. B **89** (2014) 104416 (1-14).

3. Beats and expansion of two-component Bose–Einstein condensates in the Thomas–Fermi limit: J. Q. Quach, J. Phys. B: At. Mol. Opt. Phys. **47** (2014) 215007.

4. †Selection of factorizable ground state in a frustrated spin tube: Order by disorder and hidden ferromagnetism: X. Plat, Y. Fuji, S. Capponi and P. Pujol, Phys. Rev. B **91** (2015) 064411 (1-21).

5. Distinct Trivial Phases Protected by a Point-Group Symmetry in Quantum Spin Chains: Y. Fuji, F. Pollmann and M. Oshikawa, Phys. Rev. Lett. **114** (2015) 177204.

6. Gravitational Casimir Effect: J. Q. Quach, Phys. Rev. Lett. **114** (2015) 081104 (1-5).

7. †Orbital Angular Momentum and Spectral Flow in Two-Dimensional Chiral Superfluids: Y. Tada, W. Nie and M. Oshikawa, Phys. Rev. Lett. **114** (2015) 195301.

8. 「物性物理学」～物質に潜む普遍的概念～：押川 正毅，数理科学 **620** (2015) 40-45.

9. †Absence of Quantum Time Crystals: H. Watanabe and M. Oshikawa, Phys. Rev. Lett. (2015), accepted for publication.

10. † ウラン系強磁性超伝導における強磁性ゆらぎが誘起するスピン三重項超伝導：石田 憲二，服部 泰佑，佐藤 憲昭，出口 和彦，多田 靖啓，藤本 聡，固体物理 **50** (2015) 123-132.

Tsunetsugu group

We have studied an unidentified phase transition observed in the heavy fermion compound $PrIr_2Zn_{20}$. Each Pr ion has two f-orbital electrons, and its local ground state is a non-Kramers doublet. We investigated how this degeneracy is lifted due to interactions between neighbor sites. We have performed a detailed analysis of mean-field approximation and phenomenological approach and determined the phase diagram upon controlling temperature and magnetic field. At low temperature, electric quadrupoles show a few types of antiferro orders, and the phase diagram is particularly rich when magnetic field is applied along (1,0,0) direction. An important finding is that the antiferro quadrupole order at zero magnetic field is accompanied by a weak ferro order, which is the secondary order parameter induced due to quadrupole anisotropy. Our calculation demonstrates that the induced ferro component also exhibits singular behaviors below the transition temperature, and this would explain the anomaly observed in ultrasound experiments. We have also investigated dynamics near the Mott metal-insulator transition. Using the cluster dynamical mean-field theory, we calculated dynamical correlations between doublon and holon, and demonstrated different behaviors between the metallic and insulating phases.

† Joint research with outside partners.

1. Antiferro Quadrupole Orders in Non-Kramers Doublet Systems: K. Hattori and H. Tsunetsugu, J. Phys. Soc. Jpn. **83** (2014) 034709 (19 pages).

2. Doublon dynamics of the Hubbard model on a triangular lattice: T. Sato and H. Tsunetsugu, Phys. Rev. B **90** (2014) 115114.

3. Quantum impurity in a Tomonaga-Luttinger liquid: Continuous-time quantum Monte Carlo approach: K. Hattori and A. Rosch, Phys. Rev. B **90** (2014) 115103.

Kohmoto group

Energy versus magnetic field (Hofstadter butterfly diagram) in twisted bilayer graphene is studied theoretically. If we take the usual Landau gauge, we cannot take a finite periodicity even when the magnetic flux through a supercell is a rational number. We show that the periodic Landau gauge, which has the periodicity in one direction, makes it possible to obtain the Hofstadter butterfly diagram. Since a supercell can be large, magnetic flux through a supercell normalized by the flux quantum can be a fractional number with a small denominator, even when a magnetic field is not extremely strong. As a result, quantized Hall conductance can be a solution of the Diophantine equation which cannot be obtained by the approximation of the linearized energy dispersion near the Dirac points.

Sugino group

In this fiscal year, we have studied the electrode-solution interface using the density functional theory (DFT) and the excited states of molecules using the time-dependent DFT and the two-particle Green's function method. In the former research, we focused on Pt(111)/solution interface and studies hydrogen distribution, the solvent fluctuation effect, and the oxygen reduction reaction. In the latter research, we studied the non-adiabatic effect, and in addition, started study on the firefly luciferin in collaboration with Akiyama group. The tensor-decomposition approach to very accurate first-principles calculation was also intensively studied.

1. †Effect of thermal motion on catalytic activity of nanoparticles in polar solvent: N. Bonnet, O. Sugino and M. Otani, J. Chem. Phys. **140** (2014) 044703.

2. †*First-Principles Investigation on Rydberg excitations of Firefly Luciferin Anion in Vacuum: Y. Noguchi, M. Hiyama, H. Akiyama and N. Koga, J. Chem. Phys. **141** (2014) 044309.

3. †Performance of Tamm-Dancoff approximation on nonadiabatic couplings by time-dependent density functional theory: C. Hu, O. Sugino and K. Watanabe, J. Chem. Phys. **140** (2014) 054106.

4. †Exceptionally long-ranged lattice relaxation in oxygen-deficient Ta2O5: Y. Yang, O. Sugino and Y. Kawazoe, Solid State Communications **195** (2014) 16.

5. First-principles thermodynamic description of hydrogen electroadsorption on the Pt(111) surface: T. T. T. Hanh, Y. Takimoto and O. Sugino, Surf. Sci. **625** (2014) 104.

6. †Self-Poisoning Dynamical Effects in the Oxygen Reduction Reaction on Pt(111) from a Top-Down Kinetic Analysis: N. Bonnet, M. Otani and O. Sugino, J. Phys. Chem. C **118** (2014) 13638.

7. All-electron GW+Bethe-Salpeter calculations on small molecules: D. Hirose, Y. Noguchi and O. Sugino, Phys. Rev. B **91** (2015) 205111.

8. Symmetry breaking and excitonic effects on optical properties of defective nanographenes: Y. Noguchi and O. Sugino, J. Chem. Phys. **142** (2015) 064313.

9. †TOMBO: All-electron mixed-basis approach to condensed matter physics: S. Ono, Y. Noguchi, R. Sahara, Y. Kawazoe and K. Ohno, Computer Physics Communications **189** (2015) 20.

10. *First-Principles Investigation of Strong Excitonic Effects in Oxygen 1s X-ray Absorption Spectra: Y. Noguchi, M. Hiyama, H. Akiyama, Y. Harada and N. Koga, J. Chem. Theory Comput. **11** (2015) 1668-1673.

11. *Vibronic Structures in Absorption and Fluorescence Spectra of Firefly Oxyluciferin in Aqueous Solutions: M. Hiyama, Y. Noguchi, H. Akiyama, K. Yamada and N. Koga, Photochem Photobiol (2015) published online, in print.

12. Configuration interaction with antisymmetrized geminal powers: W. Uemura, S. Kasamatsu and O. Sugino, Phys. Rev. A (2015), accepted for publication.

---

* Joint research among groups within ISSP.

Kato group

The main research subject of our laboratory is theory of nonequilibrium properties in nanoscale devices. We have performed (1) research on exact solutions for a multiorbital Anderson impurity at high bias voltages and (2) calculation of photon-assisted current noises under strong AC fields in quantum dot systems. We have also evaluated rare transition rates in evolutionary game theory in semiclassical analysis based on the path-integral formalism.

1. Dephasing in single-electron generation due to environmental noise probed by Hong-Ou-Mandel interferometry: E. Iyoda, T. Kato, K. Koshino and T. Martin, Phys. Rev. B **89** (2014) 205318(1-8).
2. Spatial effect on stochastic dynamics of bistable evolutionary games: K. H. Z. So, H. Otsuki and T. Kato, J. Stat. Mech. **-** (2014) P10020(1-34).
3. 双安定進化ゲームの確率的ダイナミクスに対する空間自由度の影響 : 曽弘 博 , 物性研究・電子版 **3** (2014) 032601(1-80).
4. メゾスコピック系の物理―基礎から最近の話題まで―（第 58 回物性若手夏の学校 : 講義）: 加藤 岳生 , 物性研究・電子版 **3** (2014) 031201(1-26).
5. Effects of Coulomb interaction on photon-assisted current noise through a quantum dot: T. J. Suzuki and T. Kato, Phys. Rev. B **91** (2015) 165302(1-12).
6. Exact Green's function for a multiorbital Anderson impurity at high bias voltages: A. Oguri and R. Sakano, Phys. Rev. B **91** (2015) 115429(1-14).

Division of Nanoscale Science

Iye group

Highly anisotropic magneto-transport is observed in single-layer graphene grown on a vicinal SiC(0001) substrate, which has a step-and-terrace morphology providing a uni-directional modulation. The anisotropy is attributed to the presence of parallel conduction due to remnant carriers in the SiC substrate which primarily conduct in the direction parallel to the steps. Well-developed quantum Hall state with zero resistance manifests itself for the current along the steps, whereas the QHE is obscured by pronounced positive magnetoresistance with quadratic magnetic-field dependence for the current across the steps. In the low field range conspicuous negative magnetoresiatance due to localization effect and oscillatory behavior attributed to geometrical resonance and observed.

1. †*Highly Anisotropic Parallel Conduction in the Stepped Substrate of Epitaxial Graphene Grown on Vicinal SiC: A. Endo, F. Komori, K. Morita, T. Kajiwara and S. Tanaka, J Low Temp Phys **179** (2015) 237-250.

Katsumoto group

By using spin-polarization at quantum point contacts (QPCs) spin interference effect in ring shaped interferometer circuit has been examined. Conductance oscillation pattern characteristic in the spin-interference effect appeared only when spin-polarized electrons were emitted into the circuit. This is probably due to the fixing of dynamical nuclear polarization. Spin injections from ferromagnets to two-dimensional electrons in quantum wells and also to two-dimensional system of graphene have been studied. In the former high spin injection efficiency of 34% has been attained. In the latter, appearance of spin-orbit interaction due to slight hydrogenaration has been detected through the inverse spin Hall effect.

1. High-efficiency graphene nanomesh magnets realized by controlling mono-hydrogenation of pore edges: T. Kato, T. Nakamura, J. Kamijyo, T. Kobayashi, Y. Yagi and J. Haruyama, Appl. Phys. Lett. **104** (2014) 252410.
2. 半導体量子輸送物性 : 勝本 信吾 , (培風館 , 東京 , 2014).
3. 量子の匠 : 勝本 信吾 , (丸善 , 東京 , 2014).

Otani group

Our research topics this year include diffusive spin transport, spin Hall effects, magnonic crystals and vortex dynamics. Concerning the diffusive spin transport, we have succeeded in fully describing spin current properties including the anisotropic spin absorption in lateral spin valves, which enable one to elucidate intrinsic spin transport and relaxation mechanisms in nonmagnets including graphene. As to spin Hall effects, we comprehensively discussed extrinsic spin Hall effects based on our

† Joint research with outside partners.

experimental results on SHE in Cu based dilute alloys. We also developed a new scheme for determining the eigenfrequency of magnetic vortex dynamics, which has been hardly detected electrically because of its nanoscale diameter on the order of the exchange length, without exploiting large magnetoresistive materials or distorted core trajectories for homodyne spin torque diode detection.

1. Propagation of nonlinearly generated harmonic spin waves in microscopic stripes: O. Rousseau, M. Yamada, K. Miura, S. Ogawa and Y. Otani, J. Appl. Phys. **115** (2014) 053914.

2. Width dependent transition of quantized spin-wave modes in Ni80Fe20 square nanorings: C. Banerjee, S. Saha, S. Barman, O. Rousseau, Y. Otani and A. Barman, J. Appl. Phys. **116** (2014) 163912.

3. Effect of anisotropic spin absorption on the Hanle effect in lateral spin valves: H. Idzuchi, Y. Fukuma, S. Takahashi, S. Maekawa and Y. Otani, Phys. Rev. B **89** (2014) 081308.

4. Extrinsic spin Hall effects measured with lateral spin valve structures: Y. Niimi, H. Suzuki, Y. Kawanishi, Y. Omori, T. Valet, A. Fert and Y. Otani, Phys. Rev. B **89** (2014) 054401.

5. Spin Injection into a Superconductor with Strong Spin-Orbit Coupling: T. Wakamura, N. Hasegawa, K. Ohnishi, Y. Niimi and Y. Otani, Phys. Rev. Lett. **112** (2014) 036602.

6. Inverse spin Hall effect in a closed loop circuit: Y. Omori, F. Auvray, T. Wakamura, Y. Niimi, A. Fert and Y. Otani, Appl. Phys. Lett. **104** (2014) 242415.

7. Modulation of effective damping constant using spin Hall effect: S. Kasai, K. Kondou, H. Sukegawa, S. Mitani, K. Tsukagoshi and Y. Otani, Appl. Phys. Lett. **104** (2014) 092408.

8. Thickness dependence of spin torque ferromagnetic resonance in Co75Fe25/Pt bilayer films: A. Ganguly, K. Kondou, H. Sukegawa, S. Mitani, S. Kasai, Y. Niimi, Y. Otani and A. Barman, Appl. Phys. Lett. **104** (2014) 072405.

9. Tunable spin wave dynamics in two-dimensional Ni80Fe20 nanodot lattices by varying dot shape: B. K. Mahato, B. Rana, D. Kumar, S. Barman, S. Sugimoto, Y. Otani and A. Barman, Appl. Phys. Lett. **105** (2014) 012406.

10. Critical exponents and domain structures of magnetic semiconductor EuS and Gd-doped EuS films near Curie temperature: H. Idzuchi, Y. Fukuma, H. S. Park, T. Matsuda, T. Tanigaki, S. Aizawa, M. Shirai, D. Shindo and Y. Otani, Appl. Phys. Express **7** (2014) 113002.

11. Detection of a symmetric circular gyration of the vortex core via the second-order harmonic magnetoresistance oscillation: S. Sugimoto, N. Hasegawa, Y. Niimi, Y. Fukuma, S. Kasai and Y. Otani, Appl. Phys. Express **7** (2014) 023006.

12. The 2014 Magnetism Roadmap: R. L. Stamps, S. Breitkreutz, J. Åkerman, A. V. Chumak, Y. Otani, G. E. W. Bauer, J.-U. Thiele, M. Bowen, S. A. Majetich, M. Kläui, I. L. Prejbeanu, B. Dieny, N. M. Dempsey and B. Hillebrands, J. Phys. D: Appl. Phys. **47** (2014) 333001.

13. Spin transport in non-magnetic nano-structures induced by non-local spin injection: H. Idzuchi, Y. Fukuma and Y. Otani, Physica E: Low-dimensional Systems and Nanostructures **68** (2015) 239.

Komori group

Electronic structures of the Au-adsorbed Ge(001) surface with a one-dimensional atomic structure were studied by high-resolution ARPES and STS. Two-dimensional Fermi surface of the surface state and decreasing density of states (DOS) toward Fermi energy are confirmed by ARPES. The latter is also confirmed by STS and attributed to the disorder of the surface lattice. The unoccupied band structure of a topological insulator BiSb alloy was studied using quasiparticle interference measured by STM. Absence of the surface band crossing is clarified, indicating this system is a dual topological insulator with the mirror Chern number −1.

1. †Systematic study of surface magnetism in Si(111)-Fe system grown by solid phase epitaxy: In situ schematic magnetic phase diagram of Si(111)-Fe: A. N. Hattori, K. Hattori, K. Kataoka, E. Takematsu, A. Ishii, F. Komori and H. Daimon, J. Magn. Magn. Mater. **363** (2014) 158-165.

2. †Absence of Luttinger liquid behavior in Au-Ge wires: A high-resolution scanning tunneling microscopy and spectroscopy study: J. Park, K. Nakatsuji, T.-H. Kim, S. K. Song, F. Komori and H. W. Yeom, Phys. Rev. B **90** (2014) 165410 (5).

3. *Robust Protection from Backscattering in the Topological Insulator $Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$: S. Kim, S. Yoshizawa, Y. Ishida, K. Eto, K. Segawa, Y. Ando, S. Shin and F. Komori, Phys. Rev. Lett. **112** (2014) 136802(1-5).

4. †*Observing hot carrier distribution in an n-type epitaxial graphene on a SiC substrate: T. Someya, H. Fukidome,

* Joint research among groups within ISSP.

Y. Ishida, R. Yoshida, T. Iimori, R. Yukawa, K. Akikubo, Sh. Yamamoto, S. Yamamoto, T. Yamamoto, T. Kanai, K. Funakubo, M. Suemitsu, J. Itatani, F. Komori, S. Shin and I. Matsuda, Appl. Phys. Lett. **104** (2014) 161103(1-4).

5. †*Scanning tunneling microscopic and spectroscopic studies on a crystalline silica monolayer epitaxially formed on hexagonal SiC(000-1) surfaces: H. Tochihara, T. Shirasawa, T. Suzuki, T. Miyamachi, T. Kajiwara, K. Yagyu, S. Yoshizawa, T. Takahashi, S. Tanaka and F. Komori, Appl. Phys. Lett. **104** (2014) 051601(1-4).

6. †*Electronic Structure and Photoelectrochemical Properties of an Ir-Doped $SrTiO_3$ Photocatalyst: S. Kawasaki, R. Takahashi, K. Akagi, J. Yoshinobu, F. Komori, K. Horiba, H. Kumigashira, K. Iwashina, A. Kudo and M. Lippmaa, J. Phys. Chem. C **118** (2014) 20222-20228.

7. †Formation of linearly linked Fe clusters on Si(111)-7x7-C_2H_5OH surface: W. Ding, D. Ju, Y. Guo, K.-I. Tanaka and F. Komori, Nanoscale Res Lett **9** (2014) 377 (6).

8. †Nonlinear terahertz field-induced carrier dynamics in photoexcited epitaxial monolayer graphene: H. A. Hafez, I. Al-Naib, M. M. Dignam, Y. Sekine, K. Oguri, F. Blanchard, D. G. Cooke, S. Tanaka, F. Komori, H. Hibino and T. Ozaki, Phys. Rev. B **91** (2015) 035422 (9).

9. *Scanning tunneling spectroscopy study of quasiparticle interference on dual topological insulator $Bi_{1-x}Sb_x$: S. Yoshizawa, F. Nakamura, A. A. Taskin, T. Iimori, K. Nakatsuji, I. Matsuda, Y. Ando and F. Komori, Phys. Rev. B **91** (2015) 045423-1,-6.

10. †*Highly Anisotropic Parallel Conduction in the Stepped Substrate of Epitaxial Graphene Grown on Vicinal SiC: A. Endo, F. Komori, K. Morita, T. Kajiwara and S. Tanaka, J Low Temp Phys **179** (2015) 237-250.

11. フェルミガス的に振る舞う表面 1 次元金属電子：矢治 浩一郎 , 小森 文夫 , 表面科学 **35** (2014) 426-431.

Yoshinobu group

We conducted several research projects in the fiscal year 2014. (1) The activation and hydrogenation of CO_2 on Cu(997) studied by SR-PES and IRAS. (2) The adsorption and decomposition of formic acid on Zn-Cu(111) studied by SR-PES and IRAS. (3) Spectroscopic characterization of chemically modified graphene (4)Construction of ambient-pressure XPS system at SPring-8. (5) The formation and characterization of oxynitride of $SrTiO_3$.

1. Quantitative analysis of chemical interaction and doping of the Si(111) native oxide surface with tetrafluorotetracyanoquinodimethane: S. Yoshimoto, M. Furuhashi, T. Koitaya, Y. Shiozawa, K. Fujimaki, Y. Harada, K. Mukai and J. Yoshinobu, J. Appl. Phys. **115** (2014) 143709 (6 pages).

2. †Configurational change of NO on Cu(110) as a function of temperature: A. Shiotari, T. Mitsui, H. Okuyama, S. Hatta, T. Aruga, T. Koitaya and J. Yoshinobu, J. Chem. Phys. **140** (2014) 214706 (6 pages).

3. Core level excitations of a fingerprint of structural and electronic properties of epitaxial silicene: R. Friedlein, A. Fleurence, K. Aoyagi, M. P. de Jong, H. Van Bui, F. B. Wiggers, S. Yoshimoto, T. Koitaya, S. Shimizu, H. Noritake, K. Mukai, J. Yoshinobu and Y. Yamada- Takamura, J. Chem. Phys. **140** (2014) 184704 (6 pages).

4. †Electronic structure of α-sexithiophene ultrathin films grown on passivated Si(001) surfaces: K. Hiraga, H. Toyoshima, H. Tanaka, K. Inoue, S. Ohno, K. Mukai, J. Yoshinobu and M. Tanaka, Appl. Surf. Sci. **307** (2014) 520-524.

5. Aqueous-Phase Oxidation of Epitaxial Graphene on the Silicon Face of SiC(0001): Md. Zakir Hossain, M. B. A. Razak, S. Yoshimoto, K. Mukai, T. Koitaya, J. Yoshinobu, H. Sone, S. Hosaka and M. C. Hersam, J. Phys. Chem. C **118** (2014) 1014-1020.

6. †*Electronic Structure and Photoelectrochemical Properties of an Ir-Doped $SrTiO_3$ Photocatalyst: S. Kawasaki, R. Takahashi, K. Akagi, J. Yoshinobu, F. Komori, K. Horiba, H. Kumigashira, K. Iwashina, A. Kudo and M. Lippmaa, J. Phys. Chem. C **118** (2014) 20222-20228.

7. †Monolayer selective methylation of epitaxial graphene on SiC(0001) through two-step chlorination-alkylation reactions: Md. Zakir Hossain, M. B. A. Razak, H. Noritake, Y. Shiozawa, S. Yoshimoto, K. Mukai, T. Koitaya, J. Yoshinobu and S. Hosaka, J. Phys. Chem. C **118** (2014) 22096-22101.

8. Structure and Photo-Induced Charge Transfer of Pyridine Molecules Adsorbed on TiO_2(110): A NEXAFS and Core-Hole-Clock Study: H. Kondoh, Y. Higashi, M. Yoshida, Y. Monya, R. Toyoshima, K. Mase, K. Amemiya, F. Tsukioka, M. Nagasaka, Y. Iwasawa, H. Orita, K. Mukai and J. Yoshinobu, Electrochemistry **82** (2014) 341-345.

9. Interface state and energy level alignment of F_4-TCNQ sandwiched between a pentacene film and the ethylene-terminated Si(100) surface: S. Yoshimoto, K. Kameshima, T. Koitaya, Y. Harada, K. Mukai and J. Yoshinobu, Organic

† Joint research with outside partners.

Electronics **15** (2014) 356-364.

10. 淡青評論「東京大学憲章を読んでみませんか」：吉信 淳 , 学内広報 **1450** (2014) 12.

11. The Quantum Nature of CH···metal Interaction: Vibrational Spectra and Kinetic and Geometric Isotope Effects of Adsorbed Cyclohexane: T. Koitaya and J. Yoshinobu, The Chemical Record **14** (2014) 848-856.

12. 「水素の事典」のうち 5 章 1-a（p.86-p.89）を分担執筆. ：水素エ ネルギー協会編 , (朝倉書店 , 東京 , 2014).

Hasegawa group

Using a low temperature scanning tunneling microscopy (LT-STM), we investigated electrical conductance at atomic point contact and found that the conductance depends on the atomic site on the substrate where the point contact is formed. For instance, the point contact conductance measured on a 3-fold hollow site of the Pb(111) substrate is larger than that measured at on-top site. We also found that the conductances measured at the two sites were reversed when the tip and substrate was separated by 20 pm from the contact. These contact-atomic-site and gap-distance dependences can be explained with the number of the substrate atoms chemically interacted with the tip-apex atom. We also studied vortices on superconducting surface structure of the Si(111)-($\sqrt{7} \times \sqrt{3}$)-In using a LT-STM. At step edges of the two-dimensional superconductor we observed vortices whose shape was elongated along the step edges and whose core still exhibited a weak superconducting gap. These anomalous features can be explained by the Josephson vortices localized at the step edges, and this also evidences the Josephson coupling at the step edges. We also studied the relation of magnetic anisotropy and stacking fault in hcp Co island structures formed on the Ag(111) substrate using a spin-polarized STM. From a magnetization curve we measured on individual islands, we found that the hcp-stacked Co islands exhibit perpendicular magnetic anisotropy whereas the ones with stacking fault have in-plane magnetization. First principle calculations clearly demonstrate the weakened magnetocrystalline anisotropy and dominance of the shape anisotropy by the stacking fault insertion into the hcp island structures.

1. †Imaging Josephson Vortices on the Surface Superconductor Si(111)-($\sqrt{7} \times \sqrt{3}$)-In using a Scanning Tunneling Microscope: S. Yoshizawa, H. Kim, T. Kawakami, Y. Nagai, T. Nakayama, X. Hu, Y. Hasegawa and T. Uchihashi, Phys. Rev. Lett. **113** (2014) 247004(1-5).

2. †Microscopic origin of the π states in epitaxial silicene: A. Fleurence, Y. Yoshida, C. -C. Lee, T. Ozaki, Y. Yamada-Takamura and Y. Hasegawa, Appl. Phys. Lett. **104** (2014) 021605 (4 pages).

3. †Scanning tunneling microscopy/spectroscopy of picene thin films formed on Ag(111): Y. Yoshida, H.-H. Yang, H.-S. Huang, S.-Y. Guan, S. Yanagisawa, T. Yokosuka, M.-T. Lin, W.-B. Su, C.-S. Chang, G. Hoffmann and Y. Hasegawa, J. Chem. Phys. **141** (2014) 114701(1-8).

4. †Excitation spectrum of Josephson vortices on surface superconductor: T. Kawakami, Y. Nagai, S. Yoshizawa, H. Kim, Y. Hasegawa, T. Nakayama, T. Uchihashi and X. Hu, J. Phys.: Conf. Ser. **568** (2014) 022022(1-5).

5. Site-Dependent Evolution of Electrical Conductance from Tunneling to Atomic Point Contact: H. Kim and Y. Hasegawa, Phys. Rev. Lett. **114** (2015) 206801(1-5).

6. †Impact of Surface Conditions on the Superconductivity of Si(111)-($\sqrt{7} \times \sqrt{3}$)-In: S. Yoshizawa, H. Kim, T. Kawakami, Y. Nagai, T. Nakayama, X. Hu, Y. Hasegawa and T. Uchihashi, e-J. Surf. Sci. Nanotech. **13** (2015) 151-154.

Lippmaa group

Our main projects are related to the development of photocatalytic oxides and nanoscale multiferroic materials. In catalyst development, we look the effects of noble metal doping in $SrTiO_3$. Recent work shows that Ir and Pt dopants tend to segregate into metallic clusters at specific oxygen activities during crystal growth. The segregated metals form nanoscale pillar structures that are embedded in the perovskite lattice and can function as embedded charge collections electrodes that help to extract photogenerated charge from the bulk of the photocatalyst. Our recent work on multiferroic materials has proven that a polar state appears in magnetite immediately at the charge ordering temperature of 120 K. A new double perovskite oxide phase, La_2NiMnO_6 was found to become multiferroic under epitaxial strain. The cause of the polar state was found to be an offset of La ions at the perovskite *A* site.

1. Growth temperature effect on the structural and magnetic properties of Fe_3O_4 films grown by the self-template method: R. Takahashi, H. Misumi and M. Lippmaa, J. Appl. Phys. **116** (2014) 033918(1-7).

2. Pulsed laser deposition of epitaxial BeO thin films on sapphire and $SrTiO_3$: T. Peltier, R. Takahashi and M. Lippmaa, Appl. Phys. Lett. **104** (2014) 231608(1-4).

3. †*Electronic Structure and Photoelectrochemical Properties of an Ir-Doped $SrTiO_3$ Photocatalyst: S. Kawasaki, R. Takahashi, K. Akagi, J. Yoshinobu, F. Komori, K. Horiba, H. Kumigashira, K. Iwashina, A. Kudo and M. Lippmaa, J.

* Joint research among groups within ISSP.

Phys. Chem. C **118** (2014) 20222-20228.

4. Spontaneous Growth of Strain-Free Magnetite Nanocrystals via Temperature-Driven Dewetting: R. Takahashi, H. Misumi, T. Yamamoto and M. Lippmaa, Crystal Growth & Design **14** (2014) 1264-1271.

5. Interfacial capacitance between a ferroelectric Fe_3O_4 thin film and a semiconducting Nb:$SrTiO_3$ substrate: R. Takahashi, Y. Cho and M. Lippmaa, J. Appl. Phys. **117** (2015) 014104(1-10).

6. A-site-driven ferroelectricity in strained ferromagnetic La_2NiMnO_6 thin films: R. Takahashi, I. Ohkubo, K. Yamauchi, M. Kitamura, Y. Sakurai, M. Oshima, T. Oguchi, Y. Cho and M. Lippmaa, Phys. Rev. B **91** (2015) 134107 (1-9).

7. †Determination of band diagram for a p-n junction between Mott insulator $LaMnO_3$ and band insulator Nb:$SrTiO_3$: M. Kitamura, M. Kobayashi, E. Sakai, R. Takahashi, M. Lippmaa, K. Horiba, H. Fujioka and H. Kumigashira, Appl. Phys. Lett. **106** (2015) 061605 (1-5).

8. †Photo-electrochemical epitaxy of silver-oxide clathrate Ag_7O_8M (M=NO_3, HSO_4) on rutile-type Nb-doped TiO_2 single crystals: R. Tanaka, R. Takahashi, S. Takata, M. Lippmaa and Y. Matsumoto, CrystEngComm **17** (2015) 3701-3707.

Division of Physics in Extreme Conditions

Uwatoko group

The electrical resistivity of the nonmagnetic quadrupolar system $PrTi_2Al_{20}$ under pressure has been measured. The experimental results suggest that critical fluctuations associated with the ferroquadrupole order may mediate superconductivity. The resistivity and ac susceptibility measurements of the topological insulator Bi_2Te_3 single crystals have been reported up to 11 GPa. Under highly hydrostatic pressure conditions generated in a multi-anvil high-pressure apparatus, a pressure- induced superconductivity only appears above a critical pressure PC ~ 7 GPa, coinciding with a structural transition from a rhombohedral to a monoclinic structure. By using an integrated-fin gasket technique made from the semi-sintered MgO ceramics and the tungsten-carbide anvils of 2.5 mm square top, we successfully generate pressures over 16 GPa at both room and cryogenic temperatures down to 0.5 K. We report on the discovery of superconductivity on the verge of antiferromagnetic order in CrAs via the application of external pressure. Bulk superconductivity with Tc≈2 K emerges at the critical pressure Pc≈8 kbar, where the first-order antiferromagnetic transition at TN≈265 K under ambient pressure is completely suppressed.

1. †*Heavy fermion superconductivity under pressure in the quadrupole system $PrTi_2Al_{20}$: K. Matsubayashi, T. Tanaka, J. Suzuki, A. Sakai, S. Nakatsuji, K. Kitagawa, Y. Kubo and Y. Uwatoko, J. Phys. Soc. Jpn. **3** (2014) 011077 (1-5).

2. †Magnetic and Fermi Surface Properties of Antiferromagnet $EuCd_{11}$: A. Nakamura, Y. Hiranaka, T. Uejo, T. Takeuchi, F. Honda, H. Harima, K. Matsubayashi, Y. Uwatoko, M. Hedo, T. Nakama and Y. Onuki, J. Phys. Soc. Jpn. **83** (2014) 074714.

3. †Superconductivity on a Crossover Phenomenon of Spin–Ladder System $SrCa_{13}Cu_{24}O_{41}$ Single Crystals: A. Hisada, K. Matsubayashi, Y. Uwatoko, N. Fujiwara, G. Deng, E. Pomjakushina, K. Conder, D. M. Radheep, R. Thiyagarajan, S. Esakkimuthu and S. Arumugam, J. Phys. Soc. Jpn. **83** (2014) 073703.

4. †Anomalous pressure dependence of the superconductivity in noncentrosymmetric LaNiC2: Evidence of strong electronic correlations: S. Katano, H. Nakagawa, K. Matsubayashi, Y. Uwatoko, H. Soeda, T. Tomita and H. Takahashi, Phys. Rev. B **90** (2014) 220508 (1-4).

5. *Doping- and pressure-induced change of electrical and magnetic properties in the Fe-based spin-ladder compound $BaFe_2Se_3$: F. Du, Y. Hirata, K. Matsubayashi, Y. Uwatoko, Y. Ueda and K. Ohgushi, Phys. Rev. B **90** (2014) 085143.

6. *High-pressure effects in anti-post-perovskite superconductors V_3PnN_x (Pn = P, As): B. S. Wang, J. -G. Cheng, K. Matsubayashi, Y. Uwatoko and K. Ohgushi, Phys. Rev. B **89** (2014) 144510 (1-4).

7. Long-range antiferromagnetic order in the frustrated XY pyrochlore antiferromagnet $Er_2Ge_2O_7$: X. Li, W. M. Li, K. Matsubayashi, Y. Sato, C. Q. Jin, Y. Uwatoko, T. Kawae, A. M. Hallas, C. R. Wiebe, A. M. Arevalo-Lopez, J. P. Attfield, J. S. Gardner, R. S. Freitas, H. D. Zhou and J. -G. Cheng, Phys. Rev. B **89** (2014) 064409 (1-7).

8. Superconductivity in the topological insulator Bi_2Te_3 under hydrostatic pressure: K. Matsubayashi, T. Terai, J. S. Zhou and Y. Uwatoko, Phys. Rev. B **90** (2014) 125126.

9. Integrated-fin gasket for palm cubic-anvil high pressure apparatus: J. -G. Cheng, K. Matsubayashi, S. Nagasaki, A. Hisada, T. Hirayama, M. Hedo, H. Kagi and Y. Uwatoko, Rev. Sci. Instrum. **85** (2014) 093907(1-6).

† Joint research with outside partners.

10. Abnormal Elastic and Vibrational Behaviors of Magnetite at High Pressures: J.-F. Lin, J. Wu, J. Zhu, Z. Mao, A. H. Said, B. M. Leu, J. Cheng, Y. Uwatoko, C. Jin and J. Zhou, Sci. Rep. **4** (2014) 6282(1-11).

11. †Magnetic Field Effect on Magnetic and Electrical Properties of$Mn_{2-x}Cu_xSb$: Y. Matsumoto, H. Orihashi, K. Matsubayashi, Y. Uwatoko, M. Hiroi and K. Koyama, IEEE Transactions on Magnetics **50** (2014) 1000704(1-4).

12. High Pressure Effect on Superconductivity of Hole-Doped Pr0.75Sr0.25FeAsO Iron Pnictides: N. R. Tamilselvan, M. Kanagaraj, K. Murata, H. Yoshino, S. Arumugam, A. Yamada, Y. Uwatoko and S. Kumararaman, J Supercond Nov Magn **27** (2014) 1381-1385.

13. Superconductivity in the vicinity of antiferromagnetic order in CrAs: W. Wu, J. Cheng, K. Matsubayashi, P. Kong, F. Lin, C. Jin, N. Wang, Y. Uwatoko and J. Luo, Nat Comms **5** (2014) 5508.

14. Correlation between T_c and Crystal Structure in S-Doped FeSe Superconductors under Pressure: Studied by X-ray Diffraction of $FeSe_{0.8}S_{0.2}$ at Low Temperatures: T. Tomita, H. Takahashi, H. Takahashi, H. Okada, Y. Mizuguchi, Y. Takano, S. Nakano, K. Matsubayashi and Y. Uwatoko, J. Phys. Soc. Jpn. **84** (2015) 024713(1-8).

15. *Temperature and composition phase diagram in the iron-based ladder compounds $Ba_{1-x}Cs_xFe_2Se_3$: T. Hawai, Y. Nambu, K. Ohgushi, F. Du, Y. Hirata, M. Avdeev, Y. Uwatoko, Y. Sekine, H. Fukazawa, J. Ma, S. Chi, Y. Ueda, H. Yoshizawa and T. J. Sato, Phys. Rev. B **91** (2015) 184416.

16. Pressure Induced Superconductivity on the border of Magnetic Order in MnP: J. -G. Cheng, K. Matsubayashi, W. Wu, J. P. Sun, F. K. Lin, J. L. Luo and Y. Uwatoko, Phys. Rev. Lett. **114** (2015) 117001(1-18).

17. Pressure-Induced Valence Crossover and Novel Metamagnetic Behavior near the Antiferromagnetic Quantum Phase Transition of $YbNi_3Ga_9$: K. Matsubayashi, T. Hirayama, T. Yamashita, S. Ohara, N. Kawamura, M. Mizumaki, N. Ishimatsu, S. Watanabe, K. Kitagawa and Y. Uwatoko, Phys. Rev. Lett. **114** (2015) 086401(1-5).

18. *High Pressure Measurements of the Resistivity of β -$YbAlB_4$: T. Tomita, K. Kuga, Y. Uwatoko and S. Nakatsuji, J. Phys.: Conf. Ser. **592** (2015) 012019.

19. Electron transport in TTF-CA under High pressures: R. Takehara, K. Miyagawa, K. Kanoda, T. Miyamoto, H. Matsuzaki, H. Okamoto, H. Taniguchi, K. Matsubayashi and Y. Uwatoko, Physica B: Condensed Matter **460** (2015) 83-87.

20. Charge disproportionation and the pressure-induced insulator–metal transition in cubic perovskite $PbCrO_3$: J. Cheng, K. E. Kweon, S. A. Larregola, Y. Ding, Y. Shirako, L. G. Marshall, Z. -Y. Li, X. Li, A. M. D. Santos, M. R. Suchomel, K. Matsubayashi, Y. Uwatoko, G. S. Hwang, J. B. Goodenough and J. -S. Zhou, Proc Natl Acad Sci USA **112** (2015) 1670-1674.

Osada group

Using a simple Slater-Koster-Harrison LCAO model, we have qualitatively studied the edge state of bilayer phosphorene, which is a unit structure of the layered crystal of black phosphorus. This model successfully reproduces the isolated edge state in the bulk gap in monolayer phosphorene. In bilayer phosphorene, however, it shows that edge states are almost buried in the valence band and there is no isolated midgap edge state at the zigzag edge. Since the buried edge state works as acceptor, holes are doped from the edge state into the bulk. This gives a possible explanation for p-type conduction in undoped black phosphorus. Under the vertical electric field, the intrinsic hole doping is reduced because a part of edge states move into the gap. These features of bilayer phosphorene might be better suited for device application.

1. 有機ディラック電子系における量子ホール状態：田嶋 尚也，佐藤 光幸，鴻池 貴子，長田 俊人，固体物理 **49** (2014) 229-240.

2. Edge State and Intrinsic Hole Doping in Bilayer Phosphorene: T. Osada, J. Phys. Soc. Jpn. **84** (2015) 013703/1-4.

3. Surface Transport in the n=0 Quantum Hall Ferromagnetic State in the Organic Dirac Fermion System: T. Osada, J. Phys. Soc. Jpn. **84** (2015) 053704/1-4.

Yamashita group

From longitudinal and transverse thermal transport measurements of kagome material Volborthite, we've found a magnon thermal conduction below the Neel temperature. We also found that there is a finite thermal Hall effect in the kagome insulator above the Neel temperature, which indicate a formation of a non-trivial spin texture as entering the QSL phase. We also have made thermal transport measurements of an organic QSL candidate, kappa-H3(CAT-EDT-TTF)2, down to 100 mK. We find that the phonons are strongly scattered in this compound down to very low temperature, although the material is very clean. By comparing the thermal conductivity of the pristine and the deuterated samples, we find the proton fluctuations in the pristine

* Joint research among groups within ISSP.

compound persist down to 300 mK. This proton fluctuation can be the origin to stabilize the QSL state of the material. As a joint research with Prof. Sasaki from IMR, Tohoku University, we've performed magnetic torque measurements at ultra-low temperatures. By using the nuclear demagnetization cryostat inherited from Ishimoto group, we've cooled organic superconductors down to 2 mK and measured the dHvA oscillations. To our best knowledge, this is the very first trial to measure a quantum oscillation below the dilution temperature. Moreover, from the temperature dependence of the torque-field curve, we succeeded to extend the vortex phase diagram of the 2D superconductor down to the ultra-low temperatures. As a joint project with Dr. Yoshida from Takigawa group in ISSP, we've also performed NMR measurements of kagome volborthite by using the ultra-low cryostat. We've found that the NMR spectrum at low fields shows a drastic change at the ultra-low temperatures, which is expected to clarify the unconventional magnetic order of the kagomé compound.

Materials Design and Characterization Laboratory

Hiroi group

A new type of magnetically frustrated lattice was found in the layered fluoride $NaBa_2Mn_3F_{11}$. A kagome-type array of regular triangles composed of Mn2+ ions (spin 5/2) deforms so as to generate the next-nearest-neighbor interaction J2 between three out of six spins in the hexagon of a normal kagome lattice, in addition to the nearest-neighbor interaction J1. As a function of |J2/J1|, this lattice can interconnect the kagome (J2 = 0) and the triangular (J2 = J1) lattices and thus is called the kagome–triangular (KT) lattice. Magnetic susceptibility and heat capacity measurements performed on a polycrystalline sample of $NaBa_2Mn_3F_{11}$ show an intensive short-range antiferromagnetic correlation below 14 K probably due to the specific magnetic frustration of the KT lattice. In addition, a long-range order at 2.0 K is observed, which is significantly low compared with the antiferromagnetic Weiss temperature of 32.3 K. Theoretical considerations of Heisenberg spins in the KT lattice reveal unique non-coplanar magnetic orders in the case of ferromagnetic J1 and antiferromagnetic J2. $NaBa_2Mn_3F_{11}$ may actually exist in this regime according to the results of our analysis based on classical Monte-Carlo simulation. In a frustrated J1–J2 chain with the nearest-neighbor ferromagnetic interaction J1 and the next-nearest-neighbor antiferromagnetic interaction J2, novel magnetic states such as a spin-nematic state are theoretically expected. However, they have been rarely examined in experiments because of the difficulty in obtaining suitable model compounds. We show here that the quasi-one-dimensional antiferromagnet $NaCuMoO_4(OH)$, which comprises edge-sharing CuO2 chains, is a good candidate J1–J2 chain antiferromagnet. The exchange interactions are estimated as J1 = −51 K and J2 = 36 K by comparing the magnetic susceptibility, heat capacity, and magnetization data with the data obtained using calculations by the exact diagonalization method. High-field magnetization measurements at 1.3 K show a saturation above 26 T with little evidence of a spin nematic state expected just below the saturation field, which is probably due to smearing effects caused by thermal fluctuations and the polycrystalline nature of the sample.

1. Novel rattling of K atoms in aluminium-doped defect pyrochlore tungstate: E. Shoko, G. J. Kearley, V. K. Peterson, H. Mutka, M. M. Koza, J.-I. Yamaura, Z. Hiroi and G. J. Thorogood, J. Phys.: Condens. Matter **26** (2014) 305401.

2. *Absence of Metallic Conductivity in Tetragonal and Cubic $PbVO_3$ at High Pressure: A. A. Belik, T. Yamauchi, H. Ueda, Y. Ueda, H. Yusa, N. Hirao and M. Azuma, J. Phys. Soc. Jpn. **83** (2014) 074711.

3. †*Electronic State of $CeFe_4As_{12}$ Investigated by Using Single Crystals Grown under High Pressure of 4 GPa: Y. Ogawa, H. Sato, M. Watanabe, T. Namiki, S. Tatsuoka, R. Higashinaka, Y. Aoki, K. Kuwahara, J.-I. Yamaura and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 034710.

4. *Kagome–Triangular Lattice Antiferromagnet $NaBa_2Mn_3F_{11}$: H. Ishikawa, T. Okubo, Y. Okamoto and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 043703(1-5).

5. †*$NaCuMoO_4(OH)$ as a Candidate Frustrated J_1–J_2 Chain Quantum Magnet: K. Nawa, Y. Okamoto, A. Matsuo, K. Kindo, Y. Kitahara, S. Yoshida, S. Ikeda, S. Hara, T. Sakurai, S. Okubo, H. Ohta and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 103702.

6. *Superconductivity in the Hypervalent Compound $Ba_2Bi(Sb_{1-x}Bi_x)_2$ with a Square-Honeycomb Lattice: T. Yajima, F. Takeiri, Y. Nozaki, Z. Li, T. Tohyama, M. A. Green, Y. Kobayashi and H. Kageyama, J. Phys. Soc. Jpn. **83** (2014) 073705.

7. Helical order and multiferroicity in the S=12 quasi-kagome system KCu3As2O7(OD)3: G. J. Nilsen, Y. Okamoto, H. Ishikawa, V. Simonet, C. V. Colin, A. Cano, L. C. Chapon, T. Hansen, H. Mutka and Z. Hiroi, Phys. Rev. B **89** (2014) 140412.

8. *Novel Phase Transitions in the Breathing Pyrochlore Lattice:^{7}Li-NMR on $LiInCr_4O_8$ and $LiGaCr_4O_8$: Y. Tanaka, M. Yoshida, M. Takigawa, Y. Okamoto and Z. Hiroi, Phys. Rev. Lett. **113** (2014) 227204(1-5).

9. †*Phase transitions and hydrogen bonding in deuterated calcium hydroxide: High-pressure and high-temperature neutron

† Joint research with outside partners.

diffraction measurements: R. Iizuka, K. Komatsu, H. Kagi, T. Nagai, A. Sano-Furukawa, T. Hattori, H. Gotou and T. Yagi, J. Solid State Chem. **218** (2014) 95.

10. †*Pressure-induced oligomerization of benzene at room temperature as a precursory reaction of amorphization: A. Shinozaki, K. Mimura, H. Kagi, K. Komatu, N. Noguchi and H. Gotou, J. Chem. Phys. **141** (2014) 084306.

11. †*Spin Polarization Measurements of Heusler Alloy $Ru_{0.2}Fe_{1.8}CrSi$ by Andreev Reflection Technique: I. Shigeta, Y. Maeda, K. Harumori, Y. Nishisako, M. Ito, T. Yamauchi and M. Hiroi, IEEE Trans. Magn. **50** (2014) 1.

12. Spin dynamics of Mn pyrochlore lattice in $YMn_2Zn_{20-x}In_x$: M. Miyazaki, R. Kadono, M. Hiraishi, I. Yamauchi, A. Koda, K. M. Kojima, I. Kawasaki, I. Watanabe, Y. Okamoto and Z. Hiroi, J. Phys.: Conf. Ser. **551** (2014) 012019.

13. Thermoelectric properties of Sr and Mg double-substituted LaCoO3 at room temperature: A. Jiamprasertboon, Y. Okamoto, Z. Hiroi and T. Siritanon, Ceramics International **40** (2014) 12729.

14. *Charge Order Induced by Cation Order in δ-$Ag_{2/3}V_2O_5$: T. Baba, T. Yamauchi, S. Yamazaki, H. Ueda, M. Isobe, Y. Matsushita and Y. Ueda, J. Phys. Soc. Jpn. **84** (2015) 024718.

15. *Superconductivity in 122-type antimonide $BaPt_2Sb_2$: M. Imai, S. Ibuka, N. Kikugawa, T. Terashima, S. Uji, T. Yajima, H. Kageyama and I. Hase, Phys. Rev. B **91** (2015) 014513.

16. Structural instability of the rutile compounds and its relevance to the metal–insulator transition of VO2: Z. Hiroi, Progress in Solid State Chemistry **43** (2015) 47.

17. *Substrate-induced anion rearrangement in epitaxial thin films of $LaSrCoO_{4-x}H_x$: G. Bouilly, T. Yajima, T. Terashima, Y. Kususe, K. Fujita, C. Tassel, T. Yamamoto, K. Tanaka, Y. Kobayashi and H. Kageyama, CrystEngComm **16** (2015) 9669-9674.

18. *Superconducting properties of $BaTi_2Pn_2O$ (Pn = Sb, Bi): T. Yajima, K. Nakano, Y. Nozaki and H. Kageyama, Physica C: Superconductivity **504** (2015) 36-38.

19. Superconductivity in LaPd2As2 with a collapsed 122 structure: S. Ganesanpotti, T. Yajima, K. Nakano, Y. Nozaki, T. Yamamoto, C. Tassel, Y. Kobayashi and H. Kageyama, Journal of Alloys and Compounds **613** (2015) 370-374.

20. Lifshitz metal–insulator transition induced by the all-in/all-out magnetic order in the pyrochlore oxide Cd2Os2O7: Z. Hiroi, J. Yamaura, T. Hirose, I. Nagashima and Y. Okamoto, APL Mater. **3** (2015) 041501.

Kawashima group

We have been investigating quantum spin/boson systems and frustrated systems by means of large-scale numerical simulation. We also develop new numerical techniques. Our group's achievements of 2014 include: (1) accumulating evidences for the deconfined critical phenomena in the SU(N) J-Q Heisenberg model, (2) proposal of new update scheme for quantum Monte Carlo method, which is based on the worm algorithm, but, unlike the conventional one, can be run by space-decomposition parallelization, and (3) large-scale non-equilibrium molecular dynamics simulation of bubble growth in under-pressured near-transition liquid.

1. *Kagome–Triangular Lattice Antiferromagnet $NaBa_2Mn_3F_{11}$: H. Ishikawa, T. Okubo, Y. Okamoto and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 043703(1-5).

2. Parallelized Quantum Monte Carlo Algorithm with Nonlocal Worm Updates: A. Masaki-Kato, T. Suzuki, K. Harada, S. Todo and N. Kawashima, Phys. Rev. Lett. **112** (2014) 140603(1-5).

3. Phase Transitions with Discrete Symmetry Breaking in Antiferromagnetic Heisenberg Models on a Triangular Lattice: R. Tamura, S. Tanaka and N. Kawashima, JPS Conf. Proc. --- Proceedings of the 12th Asia Pacific Physics Conference (APPC12) **1** (2014) 012125(1-5).

4. 大規模並列化量子モンテカルロ法の開発と冷却ボーズ原子系のシミュレーション： 正木 晶子 , スーパーコンピューティングニュース **16-6** (2014) 15-24.

5. Thermal phase transition of generalized Heisenberg models for SU(N) spins on square and honeycomb lattices: T. Suzuki, K. Harada, H. Matsuo, S. Todo and N. Kawashima, Phys. Rev. B **91** (2015) 094414.

6. Thermal phase transitions to valence-bond-solid phase in the two dimensional; generalized SU(N) Heisenberg models: T. Suzuki, K. Harada, H. Matsuo, S. Todo and N. Kawashima, J. Phys.:Conf. Ser. **592** (2015) 012114.

7. Scaling relation for dangerously irrelevant symmetry-breaking fields: T. Okubo, K. Oshikawa, H. Watanabe and N. Kawashima, Phys. Rev. B (2015), accepted for publication.

---

* Joint research among groups within ISSP.

Noguchi group

We have studied morphologies of a double-bilayer vesicle theoretically and experimentally in collaboration with Prof. M. Imai. We found that the confinement of the outer vesicle induces several shapes of the inner vesicles that had not been observed in unilamellar vesicles: double and quadruple stomatocytes, slit vesicle, and vesicles of two or three compartments with various shapes. Our simulations reproduced the experimental results very well. We also studied the assembly of banana-shaped proteins on biomembrane and dynamics of deformable microsimmers.

1. Deformable microswimmer in a swirl: Capturing and scattering dynamics: M. Tarama, A. M. Menzel and H. Löwen, Phys. Rev. E **90** (2014) 032907.
2. Morphological variation of a lipid vesicle confined in a spherical vesicle: A. Sakashita, M. Imai and H. Noguchi, Phys. Rev. E **89** (2014) 040701.
3. Tunable dynamic response of magnetic gels: Impact of structural properties and magnetic fields: M. Tarama, P. Cremer, D. Y. Borin, S. Odenbach, H. Löwen and A. M. Menzel, Phys. Rev. E **90** (2014) 042311/1-9.
4. Individual and collective dynamics of self-propelled soft particles: M. Tarama, Y. Itino, A. M. Menzel and T. Ohta, Eur. Phys. J. Spec. Top. **223** (2014) 121.
5. Multiscale modeling of blood flow: from single cells to blood rheology: D. A. Fedosov, H. Noguchi and G. Gompper, Biomech. Model. Mechanobiol. **13** (2014) 239-258.
6. Two- or three-step assembly of banana-shaped proteins coupled with shape transformation of lipid membranes: H. Noguchi, EPL **108** (2014) 48001.
7. 界面活性剤系の構造形成の粗視化分子シミュレーション : 芝隼 人 , 野口 博司 , 分子シミュレーション研究会会誌 “アンサンブル” **16** (2014) 59-65.
8. 粒子描像の流体力学計算手法 III: 野口 博司 , 分子シミュレーション研究会会誌 “アンサンブル” **16(3)** (2014) 211-214.
9. 粒子描像の流体力学計算手法 II: 野口 博司 , 分子シミュレーション研究会会誌 “アンサンブル” **16(2)** (2014) 118-121.
10. 粒子描像の流体力学計算手法 IV: 野口 博司 , 分子シミュレーション研究会会誌 “アンサンブル” **16(4)** (2014) 252-254.
11. Morphological changes of amphiphilic molecular assemblies induced by chemical reactions: K. M. Nakagawa and H. Noguchi, Soft Matter **11** (2015) 1403-1411.
12. Shape transformations of toroidal vesicles: H. Noguchi, A. Sakashita and M. Imai, Soft Matter **11** (2015) 193.

Materials Synthesis and Characterization group

1. †*Electronic State of $CeFe_4As_{12}$ Investigated by Using Single Crystals Grown under High Pressure of 4 GPa: Y. Ogawa, H. Sato, M. Watanabe, T. Namiki, S. Tatsuoka, R. Higashinaka, Y. Aoki, K. Kuwahara, J.-I. Yamaura and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 034710.
2. *Superconductivity in the Hypervalent Compound $Ba_2Bi(Sb_{1-x}Bi_x)_2$ with a Square-Honeycomb Lattice: T. Yajima, F. Takeiri, Y. Nozaki, Z. Li, T. Tohyama, M. A. Green, Y. Kobayashi and H. Kageyama, J. Phys. Soc. Jpn. **83** (2014) 073705.
3. †Magnetic properties of Mn–Bi melt-spun ribbons: T. Saito, R. Nishimura and D. Nishio-Hamane, J. Magn. Magn. Mater. **349** (2014) 9.
4. †Successive phase transitions driven by orbital ordering and electron transfer in quasi-two-dimensional $CrSe_2$ with a triangular lattice: S. Kobayashi, H. Ueda, D. Nishio-Hamane, C. Michioka and K. Yoshimura, Phys. Rev. B **89** (2014) 054413.
5. †*Phase transitions and hydrogen bonding in deuterated calcium hydroxide: High-pressure and high-temperature neutron diffraction measurements: R. Iizuka, K. Komatsu, H. Kagi, T. Nagai, A. Sano-Furukawa, T. Hattori, H. Gotou and T. Yagi, J. Solid State Chem. **218** (2014) 95.
6. †*Pressure-induced oligomerization of benzene at room temperature as a precursory reaction of amorphization: A. Shinozaki, K. Mimura, H. Kagi, K. Komatu, N. Noguchi and H. Gotou, J. Chem. Phys. **141** (2014) 084306.
7. †*Spin Polarization Measurements of Heusler Alloy $Ru_{0.2}Fe_{1.8}CrSi$ by Andreev Reflection Technique: I. Shigeta, Y. Maeda, K. Harumori, Y. Nishisako, M. Ito, T. Yamauchi and M. Hiroi, IEEE Trans. Magn. **50** (2014) 1.

† Joint research with outside partners.

8. †Magnetic properties of $SmCo_{5-x}Fe_x$ (x=0–4) melt-spun ribbon: T. Saito and D. Nishio-Hamane, J. Alloys Compd. **585** (2014) 423.

9. †Electrochemical properties of $LiMn_xFe_{1-x}PO_4$ (x = 0, 0.2, 0.4, 0.6, 0.8 and 1.0)/vapor grown carbon fiber core–sheath composite nanowire synthesized by electrospinning method: K. Kagesawa, E. Hosono, M. Okubo, D. Nishio-Hamane, T. Kudo and H. Zhou, J. Power Sources **248** (2014) 615.

10. †Electrically Conductive and Mechanically Elastic Titanium Nitride Ceramic Microsprings: S. Yang, X. Chen, K. Yamamoto, M. Iitake, D. Nishio-Hamane, H. Sakai and M. Abe, J. Nanosci. Nanotech. **14** (2014) 4292.

11. Structural discrimination of double-walled carbon nanotubes by chiral diporphyrin nanocalipers: G. Liu, Y. Saito, D. Nishio-Hamane, A. K. Bauri, E. Flahaut, T. Kimura and N. Komatsu, J. Mater. Chem. A **2** (2014) 19067.

12. Adachiite, a Si–poor member of the tourmaline supergroup from the Kiura mine, Oita Prefecture, Japan: D. Nishio–Hamane, T. Minakawa, J. Yamaura, T. Oyama, M. Ohnishi and N. Shimobayashi, J. Miner. Petrol. Sci. **109** (2014) 74.

13. Iwateite, $Na_2BaMn(PO_4)_2$, a new mineral from the Tanohata mine, Iwate Prefecture, Japan: D. Nishio-Hamane, T. Minakawa and H. Okada, J. Miner. Petrol. Sci. **109** (2014) 34.

14. Spin transition, substitution, and partitioning of iron in lower mantle minerals: K. Fujino, D. Nishio-Hamane, T. Nagai, Y. Seto, Y. Kuwayama, M. Whitaker, H. Ohfuji, T. Shinmei and T. Irifune, Phys. Earth Planet. In. **228** (2014) 186.

15. Ultrafast hydrothermal synthesis of Pr-doped $Ca_{0.6}Sr_{0.4}TiO_3$ red phosphor nanoparticles using corrosion resistant microfluidic devices with Ti-lined structure under high-temperature and high-pressure condition: K. Sue, T. Ono, Y. Hakuta, H. Takashima, D. Nishio-Hamane, T. Sato, M. Ohara, M. Aoki, Y. Takebayashi, S. Yoda, T. Hiaki and T. Furuya, Chem. Eng. J. **239** (2014) 360.

16. Magnetic properties of Sm-Fe-N bulk magnets prepared from $Sm_2Fe_{17}N_3$ melt-spun ribbons: T. Saito and D. Nishio-Hamane, J. Appl. Phys. **117** (2015) 17D130.

17. *Superconductivity in 122-type antimonide $BaPt_2Sb_2$: M. Imai, S. Ibuka, N. Kikugawa, T. Terashima, S. Uji, T. Yajima, H. Kageyama and I. Hase, Phys. Rev. B **91** (2015) 014513.

18. Effect of surfactant/water ratio and reagents' concentration on size distribution of manganese carbonate nanoparticles synthesized by microemulsion mediated route: G. Granata, F. Pagnanelli, D. Nishio-Hamane and T. Sasaki, Appl. Surf. Sci. **331** (2015) 463.

19. New hard magnetic phase in Mn–Ga–Al system alloys: T. Saito and D. Nishio-Hamane, J. Alloys Compd. **632** (2015) 486.

20. *Substrate-induced anion rearrangement in epitaxial thin films of $LaSrCoO_{4-x}H_x$: G. Bouilly, T. Yajima, T. Terashima, Y. Kususe, K. Fujita, C. Tassel, T. Yamamoto, K. Tanaka, Y. Kobayashi and H. Kageyama, CrystEngComm **16** (2015) 9669-9674.

21. *Superconducting properties of $BaTi_2Pn_2O$ (Pn = Sb, Bi): T. Yajima, K. Nakano, Y. Nozaki and H. Kageyama, Physica C: Superconductivity **504** (2015) 36-38.

22. Synthesis of oxamate and urea by oxidative single and double carbonylation of amines using immobilized palladium metal-containing ionic liquid@SBA-15: S. T. Gadge, E. N. Kusumawati, K. Harada, T. Sasaki, D. Nishio-Hamane and B. M. Bhanage, J. Mol. Catal. A: Chem. **400** (2015) 170.

23. Synthesis of Polyester Amide by Carbonylation–Polycondensation Reaction Using Immobilized Palladium Metal Containing Ionic Liquid on SBA-15 as a Phosphine-Free Catalytic System: A. Satapathy, S. T. Gadge, E. N. Kusumawati, K. Harada, T. Sasaki, D. Nishio-Hamane and B. M. Bhanage, Catal. Lett. **145** (2015) 824.

Neutron Science Laboratory

Shibayama group

Shibayama group has been exploring the structure and dynamics of soft matter, especially polymer gels, micelles, and phenolic resin, utilizing a combination of small-angle neutron scattering (SANS), neutron spin echo (NSE), and dynamic light scattering (DLS). The objectives are to elucidate the mysterious relationship between the structure and variety of novel properties/functions of polymer gels/resins. The highlights of 2013 include structural investigations of (1) Poly(ethylene glycol) in ionic liquids, (2) UCST-type phase separation of poly(N-isopropylacrylamide) in ionic liquids, (3) catalyst ink of fuel cell, (4) defect-

* Joint research among groups within ISSP.

controlled polymer networks, and so on. We also developed a dynamic light scattering microscope capable of accessing opaque samples with high spatial resolution, which sheds a light for investigations of particle dynamics and characterization in dense systems.

1. Gelation and cross-link inhomogeneity of phenolic resins studied by small- and wide-angle X-ray scattering and 1H-pulse NMR spectroscopy: A. Izumi, T. Nakao and M. Shibayama, Polymer **59** (2014) 226-233.

2. Water-in-Ionic Liquid Microemulsion Formation in Solvent Mixture of Aprotic and Protic Imidazolium-Based Ionic Liquids: T. Kusano, K. Fujii, K. Hashimoto and M. Shibayama, Langmuir **30** (2014) 11890-11896.

3. Kinetic Aspect on Gelation Mechanism of Tetra-PEG Hydrogel: K. Nishi, K. Fujii, Y. Katsumoto, T. Sakai and M. Shibayama, Macromolecules **47** (2014) 3274-3281.

4. Multiscale Dynamics of Inhomogeneity-Free Polymer Gels: T. Hiroi, M. Ohl, T. Sakai and M. Shibayama, Macromolecules **47** (2014) 763.

5. Small-Angle Neutron Scattering Study on Defect-Controlled Polymer Networks: K. Nishi, H. Asai, K. Fujii, Y.-S. Han, T.-H. Kim, T. Sakai and M. Shibayama, Macromolecules **47** (2014) 1801.

6. SANS および SAXS によるフェノール樹脂硬化物の構造解析：和泉 篤士，中尾 俊夫，岩瀬 裕希，柴山 充弘，波紋 **24** (2014) 11-14.

7. CO2 Separation Using High-toughness Ion Gel with Tetraarmed olymer Network: K. Fujii, T. Makino, K. Hashimoto, T. Sakai, M. Kanakubo and M. Shibayama, Chem. Lett. **44** (2014) 17-19.

8. Self-Oscillating Vesicles: Spontaneous Cyclic Structural Changes of Synthetic Diblock Copolymers: R. Tamate, T. Ueki, M. Shibayama and R. Yoshida, Angew. Chem. Int. Ed. **53** (2014) 11248-11252.

9. 中性子散乱を用いた構造解析手法　概説とエラストマー・高分子ゲル解析への応用：柴山 充弘，ゴム協会編 (2014) 1-10.

10. SANS studies on catalyst ink of fuel cell: M. Shibayama, T. Matsunaga, T. Kusano, K. Amemiya, N. Kobayashi and T. Yoshida, J. Appl. Polym. Sci. **131** (2014) 1-7.

11. Gelation Mechanism of Tetra-Armed Poly(ethylene glycol) in Aprotic Ionic Liquid Containing Non-volatile Proton Source, Protic Ionic Liquid: K. Hashimoto, K. Fujii, K. Nishi, T. Sakai, N. Yoshimoto, M. Morita and M. Shibayama, J. Phys. Chem. B **119** (2015) 4795-4801.

12. Dynamic light scattering study on curing mechanisms of novolac-type phenolic resins: Y. Shudo, A. Izumi, T. Takeuchi, T. Nakao and M. Shibayama, Polym. J. (2015), in print.

13. 中性子小角散乱を用いた分子集合体の解析：草野 巧巳，柴山 充弘，Colloid & Interface Communication **39** (2014) 16-18.

14. Computer simulation of network formation in natural rubber (NR): T. Nakao and S. Kohjiya, in: *Chemistry, Manufacture and Applications of Natural Rubber, Kohjiya S. and Ikeda, Y. Eds.*, edited by S. Kohjiya and Y. Ikeda, (Woodhead, Cambridge, UK, 2014), 216-246.

Yoshizawa group

A systematic study on spin dynamics in a two-dimensional transition-metal Ni oxide has been carried out with use of the high resolution chopper spectrometer installed at BL12 in the Material and Life Science Facility, J-PARC. The checkerboard-type spin-charge ordering in the highly hole-doped region of the layered nickelate was studied in detail. The nature of the excitation spectra and the thermodynamic properties in the checkerboard phase was found to show qualitative differences from those in the stripe phase. Magnetic structures and spin fluctuations in a few quantum spin systems as well as Ce-based heavy Fermion systems were also studied.

1. Magnetic structure of the conductive triangular-lattice antiferromagnet $PdCrO_2$: H. Takatsu, G. Nénert, H. Kadowaki, H. Yoshizawa, M. Enderle, S. Yonezawa, Y. Maeno, J. Kim, N. Tsuji, M. Takata, Y. Zhao, M. Green and C. Broholm, Phys. Rev. B **89** (2014) 104408.

2. Dynamical Properties of Spins and Holes in Carrier Doped Quantum Haldane Chain: T. Yokoo, S. Itoh, D. Kawana, H. Yoshizawa and J. Akimitsu, J. Phys.: Conf. Ser. **502** (2014) 012045.

3. Spin and Hole Dynamics in Carrier-Doped Quantum Haldane Chain: T. Yokoo, S. Itoh, S. Ibuka, H. Yoshizawa and J. Akimitsu, J. Phys.: Conf. Ser. **568** (2014) 042035.

4. Neutron Diffraction Study of Parasitic Nd-Moment Order in the Checkerboard-Type Phase $Nd_{1.3}Sr_{0.7}NiO_4$: R.

† Joint research with outside partners.

Kobayashi, H. Yoshizawa, M. Matsuda, R. Kajimoto, K. Ishizaka and Y. Tokura, J. Phys. Soc. Jpn. **84** (2015) 064711.

5. Resistance Anomalies Accompanying Crossover from Heavy-Fermion Regime to Intermediate-Valence Regime: A Study of Cu–Ni Substitution and Pressure Effects on $CeCu_2Si_2$: Y. Ikeda, Y. Ito, S. Araki, T. C. Kobayashi and H. Yoshizawa, J. Phys. Soc. Jpn. **84** (2015) 024702.

6. Transport and Thermodynamic Studies of Stripe and Checkerboard Ordering in Layered Nickel Oxides $R_{2-x}Sr_xNiO_4$ (R = La and Nd): Y. Ikeda, S. Suzuki, T. Nakabayashi, H. Yoshizawa, T. Yokoo and S. Itoh, J. Phys. Soc. Jpn. **84** (2015) 023706.

7. [*]Temperature and composition phase diagram in the iron-based ladder compounds $Ba_{1-x}Cs_xFe_2Se_3$: T. Hawai, Y. Nambu, K. Ohgushi, F. Du, Y. Hirata, M. Avdeev, Y. Uwatoko, Y. Sekine, H. Fukazawa, J. Ma, S. Chi, Y. Ueda, H. Yoshizawa and T. J. Sato, Phys. Rev. B **91** (2015) 184416.

8. Characterization of Ferromagnetic Order in $CePd_2P_2$: Y. Ikeda, H. Yoshizawa, S. Konishi, S. Araki, T. C. Kobayashi, T. Yokoo and S. Itoh, J. Phys.: Conf. Ser. **592** (2015) 012013.

Yamamuro group

Our laboratory is studying chemical physics of complex condensed matters by using neutron scattering, X-ray diffraction, calorimetric, dielectric, and viscoelastic techniques. Our target materials are glasses, liquids, and various disordered systems. This year, there were two important outcomes in the study of nano-particles of palladium hydrides. The neutron powder diffraction experiment of nano-PdD has demonstrated that D atoms are located not only at the octahedral sites of the fcc lattice of Pd but also at the tetrahedral sites probably due to the surface and/or distortion effects of nano-particles. The quasielastic neutron scattering experiment of nano-PdH has revealed a new relaxation, which has much smaller activation energy than that in bulk PdH. This relaxation could be due to the jump of H atoms among the tetrahedral sites. One more important topic is that our low-temperature (3 K) vapor-deposition method has produced glassy carbon dioxide that is the simplest molecular glass in the history of the glass study. Other than above topics, we have made some progresses in the studies on liquid-crystalline ionic liquids and rare-earth metal-based single molecule magnets.

1. [†]Relationship between the local dynamics and gas permeability of polyacetylenes containing polymethylated indan/tetrahydronaphtalene moieties: R. Inoue, T. Kanaya, Y. Hu, T. Masuda, K. Nishida and O. Yamamuro, Polymer **55** (2014) 182-186.

2. イミダゾリウム系イオン液体の不均一ダイナミクス：古府 麻衣子，山室 修，波紋 **24** (2014) 126-131.

3. Proton Dynamics of Two Dimensional Oxalate-Bridged Coordination Polymers: S. Miyatsu, M. Kofu, A. Nagoe, T. Yamada, M. Sadakiyo, T. Yamada, H. Kitagawa, M. Tyagi, V. Garcia Sakai and O. Yamamuro, Phys. Chem. Chem. Phys. **16** (2014) 17295-17304.

4. Thermal and Structural Studies of Imidazolium-Based Ionic Liquids with and without Liquid-Crystalline Phases: The Origin of Nanostructure: F. Nemoto, M. Kofu and O. Yamamuro, J. Phys. Chem. B **119** (2015) 5028-5034.

5. Inelastic neutron scattering study on boson peaks of imidazolium-based ionic liquids: M. Kofu, Y. Inamura, Y. Moriya, A. Podlesnyak, G. Ehlers and O. Yamamuro, J. Mol. Liq. (2015), accepted for publication.

6. 連載講座「中性子散乱による原子・分子のダイナミクスの観測」III-2　中性子非弾性散乱法による固体表面での分子分光：山室 修，古府 麻衣子，RADIOISOTOPES **63** (2014) 453-459.

7. 中性子散乱で観たイオン液体の階層的構造とダイナミクス：山室 修，根本 文也，古府 麻衣子，高圧力学会誌 **25** (2015), in print.

Masuda group

The goal of our research is to discover a new quantum phenomenon and to reveal the mechanism of it. In this fiscal year we studied the following topics; multiferroic property in integer spin systems with large single-ion anisotropy, low temperature magnetic properties of frustrated quantum spin system $Rb_2Cu_2Mo_3O_{12}$, spin gap in antiferromagnetic alternating spin 3/2 chain substances $RCrGeO_5$ (R=Y or 154Sm), and Inhomogeneous ordering in weakly coupled Heisenberg S=1/2 chains with random bonds.

1. [†*]Experimental confirmation of spin gap in antiferromagnetic alternating spin-3/2 chain substances $RCrGeO_5$(R=Y or Sm-154) by inelastic neutron scattering experiments: M. Hase, M. Soda, T. Masuda, D. Kawana, T. Yokoo, S. Itoh, A. Matsuo, K. Kindo and M. Kohno, Phys. Rev. B **90** (2014) 024416.

2. Inhomogeneous ordering in weakly coupled Heisenberg S=1/2 chains with random bonds: M. Thede, T. Haku, T.

[*] Joint research among groups within ISSP.

Masuda, C. Baines, E. Pomjakushina, G. Dhalenne, A. Revcolevschi, E. Morenzoni and A. Zheludev, Phys. Rev. B **90** (2014) 144407.

3. Spin-Nematic Interaction in the Multiferroic Compound $Ba_2CoGe_2O_7$: M. Soda, M. Matsumoto, M. Mansson, S. Ohira-Kawamura, K. Nakajima, R. Shiina and T. Masuda, Phys. Rev. Lett. **112** (2014) 127205.

4. †*Low Temperature Magnetic Properties of Frustrated Quantum Spin Chain System $Rb_2Cu_2Mo_3O_{12}$: Y. Yasui, R. Okazaki, I. Terasaki, M. Hase, M. Hagihala, T. Masuda and T. Sakakibara, JPS Conf. Proc. **3** (2014) 014014 (1-6).

5. Theoretical Study on Multiferroic Properties in Integer Spin Systems with Large Single-Ion Anisotropy: M. Matsumoto, M. Soda and T. Masuda, JPS Conf. Proc. **3** (2014) 014033.

6. Complex magnetostructural order in the frustrated spinel LiInCr4O8: G. J. Nilsen, Y. Okamoto, T. Masuda, J. Rodriguez-Carvajal, H. Mutka, T. Hansen and Z. Hiroi, Phys. Rev. B **91** (2015) 174435.

7. マルチフェロイック物質 Ba2CoGe2O7 におけるスピン・ネマティック相互作用の観測： 益田 隆嗣 , 左右田 稔 , 固体物理 **50** (2015) 111.

8. A magnetic anti-cancer compound for magnet-guided delivery and magnetic resonance imaging: H. Eguchi, M. Umemura, R. Kurotani, H. Fukumura, I. Sato, J.-H. Kim, Y. Hoshino, J. Lee, N. Amemiya, M. Sato, K. Hirata, D. J. Singh, T. Masuda, M. Yamamoto, T. Urano, K. Yoshida, K. Tanigaki, M. Yamamoto, M. Sato, S. Inoue, I. Aoki and Y. Ishikawa, Sci. Rep. **5** (2015) 9194.

International MegaGauss Science Laboratory

Takeyama group

Recently, the technique for the electro-magnetic flux compression at the International Megagauss Science Laboratory is well established and a peak magnetic field more than 700 T is reproducibly generated with the peak field almost approaching to 800 T. We have developed a calculation code, which is used for dynamical simulation of the magnetic-flux compression. This simulation assists comprehension of the time and 3 dimensional special distribution of magnetic field inside the imploding liner, and very useful for further development for achieving 1000 T. The exciton-magnon optical absorption spectra in $HgCr_2O_4$ was investigated in various magnetic phases up to the full saturation, and compared with other preceding results of the lattice and the electron-spin-resonance measurements. The existence of the novel phase, most possibly the spin nematic phase, prior to the full saturation is suggested. This phase is very similar to that discovered in other chromium spinel oxide, $CdCr_2O_4$, and $ZnCr_2O_4$. We have found that relative ordering between optical bright and dark band-edge excitons is chirality dependent in semiconductor single wall carbon nanotubes, and a bright exciton can stay at the lowest energy for a certain chirality carbon nanotube.

1. Canted 2:1:1 Magnetic Supersolid Phase in a Frustrated Magnet $MgCr_2O_4$ as a Small Limit of the Biquadratic Spin Interaction: A. Miyata, H. Ueda and S. Takeyama, J. Phys. Soc. Jpn. **83** (2014) 063702(1-4).

2. Optical Detection of Magnetic Orders in $HgCr_2O_4$ Frustrated Spin Magnet under Pulsed High Magnetic Fields: D. Nakamura, A. Miyata, Y. Aida, H. Ueda and S. Takeyama, J. Phys. Soc. Jpn. **83** (2014) 113703(1-3).

3. *Entropy of the quantum soliton lattice and multiple magnetization steps in $BiCu_2PO_6$: Y. Kohama, K. Mochidzuki, T. Terashima, A. Miyata, A. DeMuer, T. Klein, C. Marcenat, Z. L. Dun, H. Zhou, G. Li, L. Balicas, N. Abe, Y. H. Matsuda, S. Takeyama, A. Matsuo and K. Kindo, Phys. Rev. B **90** (2014) 060408(1-4).

4. Negative initial phase shift of Kerr rotation generated from the building-up process of resident electron spin polarization in a CdTe single quantum well: L. -P. Yan, M. Kurosawa, R. Kaji, G. Karczewski, S. Takeyama and S. Adachi, Phys. Rev. B **90** (2014) 205307(1-6).

5. Note: Experimental evidence of three-dimensional dynamics of an electromagnetically imploded liner: D. Nakamura, H. Sawabe and S. Takeyama, Rev. Sci. Instrum. **85** (2014) 036102.

6. †*Novel Phase of Solid Oxygen Induced by Ultrahigh Magnetic Fields: T. Nomura, Y. H. Matsuda, S. Takeyama, A. Matsuo, K. Kindo, J. L. Her and T. C. Kobayashi, Phys. Rev. Lett. **112** (2014) 247201 (5 pages).

7. †* 固体酸素の超強磁場誘起新規相 : 野村 肇宏 , 松田 康弘 , 嶽山 正二郎 , 小林 達生 , 固体物理 **Vol. 49, No. 12** (2014) 687-694.

8. †*Optical anisotropy and diamagnetic energy shifts in InP–GaP lateral quantum wells: Y. H. Shin, Y. Kim, J. D. Song, Y. T. Lee, H. Saito, D. Nakamura, Y. H. Matsuda and S. Takeyama, Journal of Luminescence **151** (2014) 244-246.

† Joint research with outside partners.

9. [*]Development of Techniques for Radio Frequency Conductivity Experiments in Ultra High Magnetic Fields: S. Lee, Y. H. Matsuda, D. Nakamura, S. Takeyama, T.-H. Arima and S. Yamada, JPS Conf. Proc. **3** (2014) 013009 (6 pages).

10. Relative Ordering between Bright and Dark Excitons in Single-walled Carbon Nanotubes: W. Zhou, D. Nakamura, H. Liu, H. Kataura and S. Takeyama, Scientific Reports **4** (2014) 6999.

11. [†*]Orbital Zeeman Effect of Liquid Oxygen in High Magnetic Fields: T. Nomura, Y. H. Matsuda, S. Takeyama, A. Matsuo, K. Kindo and T. C. Kobayashi, J. Phys. Soc. Jpn. Conf. Proc. **3** (2014) 017004(1-6).

12. [*]Magnetic-Field-Induced Insulator–Metal Transition in $(Pr_{1-y}Y_y)_{0.7}Ca_{0.3}CoO_3$ at Ultrahigh Magnetic Fields: S. Lee, Y. H. Matsuda, T. Naito, D. Nakamura and S. Takeyama, J. Phys. Soc. Jpn. **84** (2015) 044703 (5pages).

13. Exciton splitting in semiconducting carbon nanotubes in ultrahigh magnetic fields above 300 T: D. Nakamura, T. Sasaki, W. Zhou, H. Liu, H. Kataura and S. Takeyama, Phys. Rev. B **91** (2015) 235427.

14. [*]Effect of very high magnetic field on the optical properties of firefly light emitter oxyluciferin: W. Zhou, D. Nakamura, Y. Wang, T. Mochizuki, H. Akiyama and S. Takeyama, Journal of Luminescence **165** (2015) 15.

Kindo group

The heat capacity measurement by using long pulsed field with duration of about 1 sec has been opened for outer user. Newly manufactured Cu-Ag wire shows reliability and long life which enables to carry out 75 T measurement with short pulse duration.

1. [†]Almost Perfect Frustration in the Dimer Magnet $Ba_2CoSi_2O_6Cl_2$: H. Tanaka, N. Kurita, M. Okada, E. Kunihiro, Y. Shirata, K. Fujii, H. Uekusa, A. Matsuo, K. Kindo and H. Nojiri, J. Phys. Soc. Jpn. **83** (2014) 103701.

2. [†]Collapse of Anisotropic Hybridization Gap below 20 K in Kondo Semiconductor $CeFe_2Al_{10}$ by Pressure and Magnetic Field: H. Tanida, M. Nakamura, M. Sera, A. Kondo, K. Kindo, T. Nishioka and M. Matsumura, J. Phys. Soc. Jpn. **83** (2014) 084708.

3. Exchange Interactions of the Chromium Spinel Oxide $HgCr_2O_4$ in High Magnetic Fields Examined by the Magnetoelastic Theory: S. Kimura, M. Hagiwara, T. Takeuchi, H. Yamaguchi, H. Ueda and K. Kindo, J. Phys. Soc. Jpn. **83** (2014) 113709.

4. [†]Field-Induced Spin-State Transition in $LaCo_{1-x}M_xO_3$ (M = Al, Ga, Rh, and Ir): K. Sato, A. Matsuo, K. Kindo, Y. Hara, K. Nakaoka, Y. Kobayashi and K. Asai, J. Phys. Soc. Jpn. **83** (2014) 114712.

5. [†*]$NaCuMoO_4(OH)$ as a Candidate Frustrated J_1–J_2 Chain Quantum Magnet: K. Nawa, Y. Okamoto, A. Matsuo, K. Kindo, Y. Kitahara, S. Yoshida, S. Ikeda, S. Hara, T. Sakurai, S. Okubo, H. Ohta and Z. Hiroi, J. Phys. Soc. Jpn. **83** (2014) 103702.

6. [†]Spin Frustration and Field-Induced Transitions of Modified Pyrochlore Fluorides ACr_2F_6 (A = Rb and Cs): H. Ueda, A. Matsuo, K. Kindo and K. Yoshimura, J. Phys. Soc. Jpn. **83** (2014) 014701(1-6).

7. [†]Spin-Singlet Ground State of Two-Dimensional Quantum Spin Antiferromagnet $(CuCl)Ca_2Nb_3O_{10}$: Y. Tsujimoto, A. Kitada, M. Nishi, Y. Narumi, K. Kindo, T. Goko, Y. J. Uemura, A. A. Aczel, T. J. Williams, G. M. Luke, Y. Ajiro and H. Kageyama, J. Phys. Soc. Jpn. **83** (2014) 074712.

8. [†*]Successive Magnetic Transitions of $Ca_2CoSi_2O_7$ in High Magnetic Fields: M. Akaki, H. Kuwahara, A. Matsuo, K. Kindo and M. Tokunaga, J. Phys. Soc. Jpn. **83** (2014) 093704(1-4).

9. [†]Anomalous itinerant-electron metamagnetic transition in the layered $Sr_{1-x}Ca_xCo_2P_2$ system: M. Imai, C. Michioka, H. Ohta, A. Matsuo, K. Kindo, H. Ueda and K. Yoshimura, Phys. Rev. B **90** (2014) 014407.

10. [*]Entropy of the quantum soliton lattice and multiple magnetization steps in $BiCu_2PO_6$: Y. Kohama, K. Mochidzuki, T. Terashima, A. Miyata, A. DeMuer, T. Klein, C. Marcenat, Z. L. Dun, H. Zhou, G. Li, L. Balicas, N. Abe, Y. H. Matsuda, S. Takeyama, A. Matsuo and K. Kindo, Phys. Rev. B **90** (2014) 060408(1-4).

11. [†*]Experimental confirmation of spin gap in antiferromagnetic alternating spin-3/2 chain substances $RCrGeO_5$(R=Y or Sm-154) by inelastic neutron scattering experiments: M. Hase, M. Soda, T. Masuda, D. Kawana, T. Yokoo, S. Itoh, A. Matsuo, K. Kindo and M. Kohno, Phys. Rev. B **90** (2014) 024416.

12. [†]Observation of two ferromagnetic phases in Fe_3Mo_3N: T. Waki, S. Terazawa, Y. Tabata, K. Sato, A. Kondo, K. Kindo and H. Nakamura, Phys. Rev. B **90** (2014) 014416.

13. [†]Unconventional spin freezing in the highly two-dimensional spin-l/2 kagome antiferromagnet

[*] Joint research among groups within ISSP.

$Cd_2Cu_3(OH)_6(SO_4)24H_2O$: Evidence of partial order and coexisting spin singlet state on a distorted kagome lattice: M. Fujihala, X.-G. Zheng, H. Morodomi, T. Kawae, A. Matsuo, K. Kindo and I. Watanabe, Phys. Rev. B **89** (2014) 100401(1-5).

14. †*Novel Phase of Solid Oxygen Induced by Ultrahigh Magnetic Fields: T. Nomura, Y. H. Matsuda, S. Takeyama, A. Matsuo, K. Kindo, J. L. Her and T. C. Kobayashi, Phys. Rev. Lett. **112** (2014) 247201 (5 pages).

15. †High-magnetic-field phase transitions and H–T phase diagram of the Kagome-staircase compound $Ni_3V_2O_8$: Z. Q. Lin, M. Yang, H. W. Wang, Q. Guo, Y. J. Liu, X. T. Han, Y. B. Han, J. F. Wang, Z. Z. He and K. Kindo, J. Magn. Magn. Mater. **382** (2015) 7.

16. †Successive magnetic phase transitions in α-$RuCl_3$: XY-like frustrated magnet on the honeycomb lattice: Y. Kubota, H. Tanaka, T. Ono, Y. Narumi and K. Kindo, Phys. Rev. B **91** (2015) 094422.

17. †Valence-specific magnetization of the charge-ordered multiferroelectric $LuFe_2O_4$ using soft x-ray magnetic circular dichroism under 30 T pulsed high magnetic fields: Y. Narumi, T. Nakamura, K. Saito, T. Morioka, Y. Fukada, T. Kambe, N. Ikeda, Y. Kotani, T. Kinoshita, K. Kindo and H. Nojiri, Phys. Rev. B **91** (2015) 014410.

18. †*Anisotropy in the upper critical field of FeSe and $FeSe_{0.33}Te_{0.67}$ single crystals: J. L. Her, Y. Kohama, Y. H. Matsuda, K. Kindo, W.-H. Yang, D. A. Chareev, E. S. Mitrofanova, O. S. Volkova, A. N. Vasiliev and J.-Y. Lin, Supercond. Sci. Technol. **28** (2015) 045013 (6 pages).

19. †*Synchrotron X-ray spectroscopy study on the valence state and magnetization in α-$YbAl_{1-x}Fe_xB_4$ (x = 0.115) at low temperatures and high magnetic fields: T. Terashima, Y. H. Matsuda, K. Kuga, S. Suzuki, Y. Matsumoto, S. Nakatsuji, A. Kondo, K. Kindo, N. Kawamura, M. Mizumaki and T. Inami, J. Phys.: Conf. Ser. **592** (2015) 012020 (6 pages).

Tokunaga group

With using the measurement technique developed in the previous year, we studied the inverse magneto-caloric effects (IMCEs) in metamagnetic shape-memory alloys NiCoMnIn. We quantitatively estimated the contribution of magnetic, lattice, and charge degrees of freedom to the IMCEs, and revealed that the lattice sector played the dominant role in these phenomena.

1. †*Successive Magnetic Transitions of $Ca_2CoSi_2O_7$ in High Magnetic Fields: M. Akaki, H. Kuwahara, A. Matsuo, K. Kindo and M. Tokunaga, J. Phys. Soc. Jpn. **83** (2014) 093704/1-4.

2. Anisotropic Upper Critical Field of BiS_2-Based Superconductor $LaO_{0.5}F_{0.5}BiS_2$: Y. Mizuguchi, A. Miyake, K. Akiba, M. Tokunaga, J. Kajitani and O. Miura, Phys. Rev. B **89** (2014) 174515(1-7).

3. Direct measurements of inverse magnetocaloric effects in metamagnetic shape-memory alloy NiCoMnIn: T. Kihara, X. Xu, W. Ito, R. Kainuma and M. Tokunaga, Phys. Rev. B **90** (2014) 214409(1-6).

4. Novel multiferroic phase of CsCuCl3 in High Magnetic Fields: J. Shibuya, M. Akaki, Y. Kohama, A. Miyake, M. Tokunaga and H. Tanaka, J. Phys.: Conf. Ser. **568** (2014) 042030(1-5).

5. Magnetic Field-Induced Reverse Martensitic Transformation and Thermal Transformation Arrest Phenomenon of $Ni_{41}Co_9Mn_{39}Sb_{11}$ Alloy: R. Umetsu, X. Xu, W. Ito, T. Kihara, K. Takahashi, M. Tokunaga and R. Kainuma, Metals **4** (2014) 609-622.

6. Possible Excitonic Phase of Graphite in the Quantum Limit State: K. Akiba, A. Miyake, H. Yaguchi, A. Matsuo, K. Kindo and M. Tokunaga, J. Phys. Soc. Jpn. **84** (2015) 054709(1-6).

7. Magnetic control of transverse electric polarization in $BiFeO_3$: M. Tokunaga, M. Akaki, T. Ito, S. Miyahara, A. Miyake, H. Kuwahara and N. Furukawa, Nat. Commun. **6** (2015) 5878(1-5).

8. High field studies on BiFeO3 single crystals grown by the laser-diode heating floating zone method: M. Tokunaga, M. Akaki, A. Miyake, T. Ito and H. Kuwahara, Journal of Magnetism and Magnetic Materials **383** (2015) 259-261.

Y. Matsuda group

A novel phase of solid oxygen was discovered in ultrahigh magnetic fields of up to 193 T. The phase transition is expected to be driven by rearrangement of oxygen molecules. The nearest neighbor molecules are coupled with the parallel H-geometry at low magnetic fields and the exchange interaction is antiferromagnetic. The H-geometry becomes unstable at high magnetic fields owing to its antiferromagnetic exchange interaction and a new geometry is likely to realize so that the exchange coupling becomes ferromagnetic. The cubic crystal symmetry is most plausible for the novel phase that we term θ phase. Other than solid oxygen, heavy fermion compounds such as $YbAlB_4$ and $CeRh_2Si_2$ were investigated in pulsed high magnetic fields. A

† Joint research with outside partners.

small metamagnetic transition was observed in $YbAlB_4$ and the origin of the transition can be the valence change of Yb. As for $CeRh_2Si_2$, a field-induced valence change of Ce at around 25 T was directly observed by X-ray absorption spectroscopy done at SPring-8. In order to realize the electrical transport measurement in destructive magnetic fields over 100 T, a contactless radio frequency technique was developed. Magnetic-field-induced insulator-metal transition of $Pr_{0.6}Ca_{0.4}MnO_3$ was successfully observed in pulsed fields up to 80 T produced by the single-turn coil method.

1. *Entropy of the quantum soliton lattice and multiple magnetization steps in $BiCu_2PO_6$: Y. Kohama, K. Mochidzuki, T. Terashima, A. Miyata, A. DeMuer, T. Klein, C. Marcenat, Z. L. Dun, H. Zhou, G. Li, L. Balicas, N. Abe, Y. H. Matsuda, S. Takeyama, A. Matsuo and K. Kindo, Phys. Rev. B **90** (2014) 060408(1-4).

2. †*Novel Phase of Solid Oxygen Induced by Ultrahigh Magnetic Fields: T. Nomura, Y. H. Matsuda, S. Takeyama, A. Matsuo, K. Kindo, J. L. Her and T. C. Kobayashi, Phys. Rev. Lett. **112** (2014) 247201 (5 pages).

3. †* 固体酸素の超強磁場誘起新規相：野村 肇宏 , 松田 康弘 , 嶽山 正二郎 , 小林 達生 , 固体物理 **Vol. 49, No. 12** (2014) 687-694.

4. †*Optical anisotropy and diamagnetic energy shifts in InP–GaP lateral quantum wells: Y. H. Shin, Y. Kim, J. D. Song, Y. T. Lee, H. Saito, D. Nakamura, Y. H. Matsuda and S. Takeyama, Journal of Luminescence **151** (2014) 244-246.

5. *Development of Techniques for Radio Frequency Conductivity Experiments in Ultra High Magnetic Fields: S. Lee, Y. H. Matsuda, D. Nakamura, S. Takeyama, T.-H. Arima and S. Yamada, JPS Conf. Proc. **3** (2014) 013009 (6 pages).

6. Valence State in Ce-based Heavy Fermion Compounds at High Magnetic Fields: Y. H. Matsuda, J.-L. Her, S. Michimura, T. Inami, T. Ebihara and H. Amitsuka, JPS Conf. Proc. **3** (2014) 011044 (6 pages).

7. †*Orbital Zeeman Effect of Liquid Oxygen in High Magnetic Fields: T. Nomura, Y. H. Matsuda, S. Takeyama, A. Matsuo, K. Kindo and T. C. Kobayashi, J. Phys. Soc. Jpn. Conf. Proc. **3** (2014) 017004(1-6).

8. *Magnetization of Yb-based mixed-valent compounds at megagauss fields: T. Terashima, Y. H. Matsuda, K. Kuga, Y. Matsumoto and S. Nakatsuji, J. Phys. Conf. Proc. **3** (2014) 011027(1-5).

9. *Magnetic-Field-Induced Insulator–Metal Transition in $(Pr_{1-y}Y_y)_{0.7}Ca_{0.3}CoO_3$ at Ultrahigh Magnetic Fields: S. Lee, Y. H. Matsuda, T. Naito, D. Nakamura and S. Takeyama, J. Phys. Soc. Jpn. **84** (2015) 044703 (5pages).

10. †*Anisotropy in the upper critical field of FeSe and $FeSe_{0.33}Te_{0.67}$ single crystals: J. L. Her, Y. Kohama, Y. H. Matsuda, K. Kindo, W.-H. Yang, D. A. Chareev, E. S. Mitrofanova, O. S. Volkova, A. N. Vasiliev and J.-Y. Lin, Supercond. Sci. Technol. **28** (2015) 045013 (6 pages).

11. † 超強磁場で誘起される固体酸素の新しい相：松田 康弘 , パリティ **30** (2015) 48-51.

12. †*Synchrotron X-ray spectroscopy study on the valence state and magnetization in α-$YbAl_{1-x}Fe_xB_4$ (x = 0.115) at low temperatures and high magnetic fields: T. Terashima, Y. H. Matsuda, K. Kuga, S. Suzuki, Y. Matsumoto, S. Nakatsuji, A. Kondo, K. Kindo, N. Kawamura, M. Mizumaki and T. Inami, J. Phys.: Conf. Ser. **592** (2015) 012020 (6 pages).

Center of Computational Materials Science

Akai group

(1) In order to seek better permanent magnet materials, the mechanisms that govern the magnetic anisotropy, magnetisation, and the Curie temperature were investigated. For typical permanent magnet materials $Nd_2Fe_{17}B$ and $Sm_2Fe_{17}N_3$, the local magnetic moments, hyperfine fields, the magnetic exchange couplings, the magnetic anisotropy energy are calculated within the density functional theory using all-eletron KKR Green's function method. The importance of hybridisation between the 4f states of rare earth elements and N and B are pointed out. (2)New scheme of calculating Keldysh Green's function in the framework of KKR Green's function was proposed and successfully applied to some test cases. The method open up a possibility to deal with non-equlibrium process in the framework of KKR all -electron scheme. (3)Extension of the optimised effective potential method (OEP) so as to include the correlation of RPA level was developed together with some reformulation of OEP that as it stood had shown ill behaviour for spin polarised cases.

1. Ab initio study of ^{59}Co NMR spectra in $Co_2FeAl_{1-x}Si_x$ Heusler alloys: H. Nishihara, K. Sato, H. Akai, C. Takiguchi, M. Geshi, T. Kanomata, T. Sakon and T. Wada, Physica B **485** (2015) 66-70.

2. Optimized effective potential method and application to a static RPA correlation: T. Fukazawa and H. Akai, J. Phys.: Condens. Matter **27** (2015) 115502(1-10).

* Joint research among groups within ISSP.

3. Formulation of the augmented plane-wave and muffin-tin orbital method: T. Kotani, H. Kino and H. Akai, J. Phys. Soc. Jpn. **84** (2015) 034701(1-9(.

4. The metamagnetic behavior and giant inverse magnetocaloric effect in Ni–Co–Mn– (Ga, In, Sn) Heusler alloys: P. Entel, V. V. Sokolovskiy, V. D. Buchelnikov, M. Ogura, M. E. Gruner, A. Gruenebohm, D. Comtesse and H. Akai, J. Magn. Magn. Mater. **385** (2015) 193-197.

5. Role of N in permeant magnet materials $Sm_2Fe_{17}N_x$: M. Ogura, A. Mashiyama and H. Akai, J. Phys. Soc. Jpn. (2015), in print.

6. Basic and applications of Moessbauer spectrometry and the electronics structure of matters: H. Akai, ISOTOPES **63** (2014) 163-174.

7. 鉄の事典：赤井 久純 , (朝倉書店 , 東京 , 2014).

Ozaki group

Although it is believed that two-dimensional honeycomb structures consisting of silicon atoms do not exist experimentally due to relative instability of its hybridized sp_2 orbitals, a recent experiment clearly demonstrates that silicene, honeycomb structure of silicon atoms, can be fabricated on ZrB_2 (0001) thin films. We have performed detailed studies on geometrical and electronic structures of silicene on ZrB_2 and a related two-dimensional structure by means of electronic structure calculations based on density functional theories (DFT), guided by a close collaboration with experimental groups. Theoretical chemical shift of Si-2p states and band structure calculations strongly support the formation of silicene having a planar-like structure. The stability of the planar-like structure over the regularly buckled structure can be understood by interaction between states of the silicene and surface states consisting of the d-orbital of the top Zr atoms. We have also proposed a possible mechanism for the formation of the domain structure of silicene on ZrB_2. It is inferred that the domain structure is induced by an instability of a phonon having a nearly zero frequency, and is formed in such a way that the k-points having the zero frequency can be removed from the first Brillouin zone. The mechanism is verified by performing large-scale total energy calculations. We have further explored a possible structure of multi-layer silicene, and find that the MoS_2 structure consisting of silicon atoms is stabilized with atoms in the inner layer having a sixfold coordination, which results in cigar-shaped nematic orbitals originating from the Si-sp_2 orbitals.

1. Systematic study of electronic and magnetic properties for $Cu_{12-x}TM_xSb_4S_{13}$ (TM= Mn, Fe, Co, Ni, and Zn) tetrahedrite: K. Suekuni, Y. Tomizawa, T. Ozaki and M. Koyano, J. Appl. Phys. **115** (2014) 143702.

2. Avoiding critical-point phonon instabilities in two-dimensional materials: The origin of the stripe formation in epitaxial silicene: C. -C. Lee, A. Fleurence, R. Friedlein, Y. Yamada-Takamura and T. Ozaki, Phys. Rev. B **90** (2014) 241402(R).

3. Band structure of silicene on zirconium diboride (0001) thin-film surface: Convergence of experiment and calculations in the one-Si-atom Brillouin zone: C. -C. Lee, A. Fleurence, Y. Yamada-Takamura, T. Ozaki and R. Friedlein, Phys. Rev. B **90** (2014) 075422.

4. Competing magnetism in π-electrons in graphene with a single carbon vacancy: C. -C. Lee, Y. Yamada-Takamura and T. Ozaki, Phys. Rev. B **90** (2014) 014401.

5. Diverse forms of bonding in two-dimensional Si allotropes: Nematic orbitals in the MoS_2 structure: F. Gimbert, C. -C. Lee, R. Friedlein, A. Fleurence, Y. Yamada-Takamura and T. Ozaki, Phys. Rev. B **90** (2014) 165423.

6. Microscopic origin of the π states in epitaxial silicene: A. Fleurence, Y. Yoshida, C. -C. Lee, T. Ozaki, Y. Yamada-Takamura and Y. Hasegawa, Appl. Phys. Lett. **104** (2014) 021605.

7. Strain effects on the magnetic anisotropy of $Y_2Fe_{14}B$ examined by first-principles calculations: Z. Torbatian, T. Ozaki, S. Tsuneyuki and Y. Gohda, Appl. Phys. Lett. **104** (2014) 242403.

8. A method of orbital analysis for large-scale first-principles simulations: T. Ohwaki, M. Otani and T. Ozaki, J. Chem. Phys. **140** (2014) 244105.

9. First-principles electronic transport calculations of graphene nanoribbons on SiO_2/Si: H. Jippo, T. Ozaki and M. Ohfuchi, Appl. Phys. Express **7** (2014) 025101.

10. A decomposition method with minimum communication amount for parallelization of multi-dimensional FFTs: T. V. T. Duy and T. Ozaki, Comput. Phys. Commun. **185** (2014) 153.

11. A three-dimensional domain decomposition method for large-scale DFT electronic structure calculations: T. V. T. Duy and T. Ozaki, Comput. Phys. Commun. **185** (2014) 777.

† Joint research with outside partners.

Laser and Synchrotron Research Center

Suemoto group

(1) Coherent control and observation of spin dynamics by using terahertz (THz) wave: The spin orientation during the spin reorientation transition was precisely determined in an orthoferrite $Dy_{0.7}Er_{0.3}FeO_3$ by THz TDS (time domain spectroscopy) and the advantage of this method over a standard SQUID measurement was demonstrated. By using a high intensity THz source, instantaneous non-linear response was found in $ErFeO_3$. (2) THz spectroscopy for d-electron dynamics: Behavior of photo-generated d-electrons in MnO was investigated by optical-pump-THz-probe spectroscopy and the relaxation path to magnetic self-trapped exciton was suggested, comparing with luminescence measurement. (3) Ultrafast time-resolved luminescence spectroscopy: Luminescence dynamics was studied in graphenes and the importance of Fermi energy to the hot electron relaxation was shown. Femtosecond infrared luminescence was firstly observed in semimetals (Bi, Sb) and in a topological insulator ($TlBiSe_2$). (4) Single-shot soft x-ray imaging: A single-shot soft-x-ray microscope with a Fresnel's zone plate was constructed and a good performance with a spatial resolution of 200 nm was demonstrated.

1. †Enhanced spin-precession dynamics in a spin-metamaterial coupled resonator observed in terahertz time-domain measurements: T. Kurihara, K. Nakamura, K. Yamaguchi, Y. Sekine, Y. Saito, M. Nakajima, K. Oto, H. Watanabe and T. Suemoto, Phys. Rev. B **90** (2014) 144408(1-5).

2. †Nonlinear photoluminescence properties of trions in hole-doped single-walled carbon nanotubes: N. Akizuki, M. Iwamura, S. Mouri, Y. Miyauchi, T. Kawasaki, H. Watanabe, T. Suemoto, K. Watanabe, K. Asano and K. Matsuda, Phys. Rev. B **89** (2014) 195432.

3. †Ultrafast dynamics of photoinduced semiconductor-to-metal transition in optical switching nano-oxide Ti_3O_5: A. Asahara, H. Watanabe, H. Tokoro, S. Ohkoshi and T. Suemoto, Phys. Rev. B **90** (2014) 014303(1-7).

4. *Gain-switching dynamics in optically pumped single-mode InGaN vertical-cavity surface-emitting lasers: S. Chen, A. Asahara, T. Ito, J. Zhang, B. Zhang, T. Suemoto, M. Yoshita and H. Akiyama, Optics Express **22** (2014) 4196-4201.

5. †Domain growth and aggregation dynamics in photo-induced phase transition phenomena: H. Watanabe, S. Ishige, A. Asahara, H. Tokoro, S. Ohkoshi and T. Suemoto, J. Lumin **152** (2014) 108-111.

6. *Direct generation of 2-ps blue pulses from gain-switched InGaN VCSEL assessed by up-conversion technique: A. Asahara, S. Chen, T. Ito, M. Yoshita, W. Liu, B. Zhang, T. Suemoto and H. Akiyama, Sci. Rep. **4** (2014) 6401.

7. *Spectral dynamics of picosecond gain-switched pulses from nitride-based vertical-cavity surface-emitting lasers: S. Chen, T. Ito, A. Asahara, M. Yoshita, W. Liu, J. Zhang, B. Zhang, T. Suemoto and H. Akiyama, Sci. Rep. **4** (2014) 4325.

8. †Submicron scale image observation with a grazing incidence reflection-type single-shot soft X-ray microscope: M. Baba, M. Nishikino, N. Hasegawa, T. Tomita, Y. Minami, R. Takei, M. Yamagiwa, T. Kawachi and T. Suemoto, Jpn J. App. Phys. **53** (2014) 080302.

9. †Time-Resolved Soft X-Ray Imaging of Femtosecond Laser Ablation Processes on MetalsTomita: M. Nishikino, N. Hasegawa, Y. Minami, R. Takei, M. Baba, T. Eyama, S. Takayoshi, T. Kaihori, T. Morita, Y. Hirano, T. Kawachi, M. Yamagiwa and T. Suemoto, J. Laser Micro Nanoengineering **9** (2014) 137-142.

10. †High-frequency millimeter wave absorption of indium-substituted epsilon-Fe_2O_3 spherical nanoparticles: M. Yoshikiyo, A. Namai, M. Nakajima, K. Yamaguchi, T. Suemoto and S. Ohkoshi, J. App. Phys. **15** (2014) 172613.

11. †Ultraviolet stimulated emission from high-temperature-annealed MgO microcrystals at room temperature: H. Soma, Y. Uenaka, A. Asahara, T. Suemoto and T. Uchino, Appl. Phys. Lett. **106** (2015) 041116(1-5).

Shin group

We studied high Tc Fe-pnictide superconductors using 7-eV laser. High resolution photoemission study with polarization dependence is very powerful for the study of the superconducting mechanism. Orbital fluctuation mechanism is also important in addition to the spin fluctuation mechanism.

1. Electronic structure of CeCuAs2: A. Chainani, M. Matsunami, M. Taguchi, R. Eguchi, Y. Takata, M. Oura, S. Shin, K. Sengupta, E. V. Sampathkumaran, Th. Doert, Y. Senba, H. Ohashi, K. Tamasaku, Y. Kohmura, M. Yabashi and T. Ishikawa, Phys. Rev. B **89** (2014) 235117.

2. *Evidence for excluding the possibility of d-wave superconducting-gap symmetry in Ba-doped KFe_2As_2: Y. Ota, K. Okazaki, Y. Kotani, T. Shimojima, W. Malaeb, S. Watanabe, C. -T. Chen, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, T. Saito, H. Fukazawa, Y. Kohori and S. Shin, Phys. Rev. B **89** (2014) 081103(1-5).

---

* Joint research among groups within ISSP.

3. †*Evidence of a universal relation between electron-mode coupling and T_c in $Ba_{1-x}K_xFe_2As_2$ superconductor from laser angle-resolved photoemission spectroscopy: W. Malaeb, T. Shimojima, Y. Ishida, T. Kondo, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, T. Saito, Y. Kohori and S. Shin, Phys. Rev. B **90** (2014) 195124(1-6).

4. †*Observation of a giant Kerr rotation in a ferromagnetic transition metal by M-edge resonant magneto-optic Kerr effect: Sh. Yamamoto, M. Taguchi, M. Fujisawa, R. Hobara, S. Yamamoto, K. Yaji, T. Nakamura, K. Fujikawa, R. Yukawa, T. Togashi, M. Yabashi, M. Tsunoda, S. Shin and I. Matsuda, Phys. Rev. B **89** (2014) 064423(1-6).

5. *Pseudogap formation above the superconducting dome in iron pnictides: T. Shimojima, T. Sonobe, W. Malaeb, K. Shinada, A. Chainani, S. Shin, T. Yoshida, S. Ideta, A. Fujimori, H. Kumigashira, K. Ono, Y. Nakashima, H. Anzai, M. Arita, A. Ino, H. Namatame, M. Taniguchi, M. Nakajima, S. Uchida, Y. Tomioka, T. Ito, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, K. Ohgushi, S. Kasahara, T. Terashima, H. Ikeda, T. Shibauchi, Y. Matsuda and K. Ishizaka, Phys. Rev. B **89** (2014) 045101(1-10).

6. †*Ultrafast photoinduced transition of an insulating VO_2 thin film into a nonrutile metallic state: R. Yoshida, T. Yamamoto, Y. Ishida, H. Nagao, T. Otsuka, K. Saeki, Y. Muraoka, R. Eguchi, K. Ishizaka, T. Kiss, S. Watanabe, T. Kanai, J. Itatani and S. Shin, Phys. Rev. B **89** (2014) 205114(1-7).

7. *Time-resolved photoemission apparatus achieving sub-20-meV energy resolution and high stability: Y. Ishida, T. Togashi, K. Yamamoto, M. Tanaka, T. Kiss, T. Otsu, Y. Kobayashi and S. Shin, Rev. Sci. Instrum. **85** (2014) 123904(1-9).

8. *Robust Protection from Backscattering in the Topological Insulator $Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$: S. Kim, S. Yoshizawa, Y. Ishida, K. Eto, K. Segawa, Y. Ando, S. Shin and F. Komori, Phys. Rev. Lett. **112** (2014) 136802(1-5).

9. Selective Probing of the OH or OD Stretch Vibration in Liquid Water Using Resonant Inelastic Soft-X-Ray Scattering: Y. Harada, T. Tokushima, Y. Horikawa, O. Takahashi, H. Niwa, M. Kobayashi, M. Oshima, Y. Senba, H. Ohashi, KT. Wikfeldt, A. Nilsson, LGM. Pettersson and S. Shin, Phys. Rev. Lett. **111** (2014) 193001(1-5).

10. レーザー光電子分光による分子性導体の電子構造の観測：石坂 香子，小泉 健二，木須 孝幸，辛 埴，固体物理 **49** (2014) 153-162.

11. †*Observing hot carrier distribution in an n-type epitaxial graphene on a SiC substrate: T. Someya, H. Fukidome, Y. Ishida, R. Yoshida, T. Iimori, R. Yukawa, K. Akikubo, Sh. Yamamoto, S. Yamamoto, T. Yamamoto, T. Kanai, K. Funakubo, M. Suemitsu, J. Itatani, F. Komori, S. Shin and I. Matsuda, Appl. Phys. Lett. **104** (2014) 161103(1-4).

12. †*Solvation dependence of valence electronic states of water diluted in organic solvents probed by soft X-ray spectroscopy: T. Tokushima, Y. Horikawa, O. Takahashi, H. Arai, K. Sadakane, Y. Harada, Y. Takata and S. Shin, Phys. Chem. Chem. Phys. **16** (2014) 10753-10761.

13. *Superconductivity in an electron band just above the Fermi level: possible route to BCS-BEC superconductivity: K. Okazaki1, Y. Ito, Y. Ota, Y. Kotani, T. Shimojima, T. Kiss, S. Watanabe, C. -T. Chen, S. Niitaka, T. Hanaguri, H. Takagi, A. Chainani and S. Shin, Sci. Rep. **4** (2014) 4109(1-6).

14. Development of a single-shot CCD-based data acquisition system for time-resolved X-ray photoelectron spectroscopy at an X-ray free-electron laser facility: M. Oura, T. Wagai, A. Chainani, J. Miyawaki, H. Sato, M. Matsunami, R. Eguchi, T. Kiss, T. Yamaguchi, Y. Nakatani, T. Togashi, T. Katayama, K. Ogawa, M. Yabashi, I. Y. Tanaka, Y. Kohmura, K. Tamasaku, S. Shin and T. Ishikawa, J.Syn.Rad. **21** (2014) 183-192.

15. The electronic structure of carbonate ion in aqueous solution studied by soft X-ray emission spectroscopy: Y. Horikawa A. Yoshida, O. Takahashi, H. Araia, T. Tokushima, T. Gejo and S. Shin, J.Mol.Liq **189** (2014) 9-12.

16. †*New soft X-ray beamline BL07LSU at SPring-8: S. Yamamoto, Y. Senba, T. Tanaka, H. Ohashi, T. Hirono, H. Kimura, M. Fujisawa, J. Miyawaki, A. Harasawa, T. Seike, S. Takahashi, N. Nariyama, T. Matsushita, M. Takeuchi, T. Ohata, Y. Furukawa, K. Takeshita, S. Goto, Y. Harada, S. Shin, H. Kitamura, A. Kakizaki, M. Oshima and I. Matsuda, J Synchrotron Rad **21** (2014) 352-365.

17. Observation of quadrupole helix chirality and its domain structure in DyFe3(BO3)4: T. Usui, Y. Tanaka, H. Nakajima, M. Taguchi, A. Chainani, M. Oura, S. Shin, N. Katayama, H. Sawa, Y. Wakabayashi and T. Kimura, Nature materials **13** (2014) 611-618.

18. Ultrahigh-spatial-resolution chemical and magnetic imaging by laser-based photoemission electron microscopy: T. taniuchi, Y. Kotani and S. Shin, Rev. Sci. Instrum. **86** (2015) 023701.

19. *Emergent photovoltage on SmB_6 surface upon bulk-gap evolution revealed by pump-and-probe photoemission spectroscopy: Y. Ishida, T. Otsu, T. Shimada, M. Okawa, Y. Kobayashi, F. Iga, Y. Takabatake and S. Shin, Sci. Rep. **5**

† Joint research with outside partners.

(2015) 8160(1-6).

20. * 鉄タンパク質の電子状態と機能：原田 慈久 , 辛 埴 , (朝倉書店「鉄の事典」, 3.11.d 節 , 2014).

Takahashi group

We have been studying the structure and phase transition of surfaces and interfaces with diffraction techniques. Atomic structure and electronic transport properties of Cu-doped topological insulator Bi_2Se_3 thin-films were investigated by surface X-ray diffraction and magnetotransport measurements. It is reported that Cu-doped Bi_2Se_3 bulk crystals show superconductivity at low temperature below 4 K. But the thin-film samples never showed superconductivity. This suggests that such bulk crystals contain impurity sub-phases responsible to superconductivity. On the other hand, we investigated the surface structure of Si(111)-5x2-Au, a typical prototype of one dimensional structure, that has not been solved for a long time since its first observation by LEED in 1969. We have experimentally determined the atomic structure without ambiguity with an aid of a holographic method in surface X-ray diffraction.

1. †Structure and transport properties of Cu doped Bi_2Se_3 films: T. Shirasawa, M. Sugiki, T. Hirahara, M. Aitani, T. Shirai, S. Hasegawa and T. Takahashi, Phys. Rev. B **89** (2014) 195311(1-6).

2. †Structure determination of multilayer silicene grown on Ag(111) films by electron diffraction: Evidence for Ag segregation at the surface: T. Shirai, T. Shirasawa, T. Hirahara, N. Fukui, T. Takahashi and S. Hasegawa, Phys. Rev. B **89** (2014) 241403(R)(1-5).

3. Identification of the Structure Model of the Si(111)-(5×2)-Au Surface: T. Shirasawa, W. Voegeli, T. Nojima, Y. Iwasawa, Y. Yamaguchi and T. Takahashi, Phys. Rev. Lett. **113** (2014) 165501(1-4).

4. †*Scanning tunneling microscopic and spectroscopic studies on a crystalline silica monolayer epitaxially formed on hexagonal SiC(000-1) surfaces: H. Tochihara, T. Shirasawa, T. Suzuki, T. Miyamachi, T. Kajiwara, K. Yagyu, S. Yoshizawa, T. Takahashi, S. Tanaka and F. Komori, Appl. Phys. Lett. **104** (2014) 051601(1-4).

5. †Determination of atomic positions in silicene on Ag(111) by low-energy electron diffraction: K. Kawahara, T. Shirasawa, R. Arafune, C. -L. Lin, T. Takahashi, M. Kawai and N. Takagi, Surf. Sci. **623** (2014) 25-28.

Akiyama group

In 2014, we developed experimental study on subcells diagnosis in a multi-junction solar cell via absolute electroluminescence-efficiency measurements, on the basis of a detailed-balance or reciprocity relation. Collaborations on GaAs quantum wells pumped by an intense single-cycle terahertz pulse were also developed. We studied spectral dynamics in pico-second short-pulse generation via gain switching in InGaN VCSELs and double-core-slab-waveguide semiconductor lasers for end optical pumping. We studied temperature dependence of quantitative green and orange/red emission intensities in firefly bioluminescence, and made intensive studies on theoretical quantum-chemistry calculations on luciferins and oxyluciferins.

1. Mode imaging and loss evaluation of semiconductor waveguides: T. Mochizuki, C. Kim, M. Yoshita, T. Nakamura, H. Akiyama, L. N. Pfeiffer and K. W. West, Rev. Sci. Instrum. **85** (2014) 053109.

2. Impact of Sub-cell Internal Luminescence Yields on Energy Conversion Efficiencies of Tandem Solar Cells: A design principle: L. Zhu, C. Kim, M. Yoshita, S. Chen, S. Sato, T. Mochizuki, H. Akiyama and Y. Kanemitsu, Appl. Phys. Lett. **104** (2014) 031118.

3. Robust red-emission spectra and yields in firefly bioluminescence against temperature changes: T. Mochizuki, Y. Wang, M. Hiyama and H. Akiyama, Appl. Phys. Lett. **104** (2014) 213704.

4. Spectroscopic Study of Firefly Oxyluciferin in an Enzymatic Environment on the Basis of Stability Monitoring: Y. Wang, Y. Hayamizu and H. Akiyama, J. Phys. Chem. B **118** (2014) 2070–2076.

5. †*First-Principles Investigation on Rydberg excitations of Firefly Luciferin Anion in Vacuum: Y. Noguchi, M. Hiyama, H. Akiyama and N. Koga, J. Chem. Phys. **141** (2014) 044309.

6. ホタル生物発光と物理：秋山 英文 , 望月 敏光 , 樋山 みやび , 日本物理学会誌 **69** (2014) 218.

7. *Gain-switching dynamics in optically pumped single-mode InGaN vertical-cavity surface-emitting lasers: S. Chen, A. Asahara, T. Ito, J. Zhang, B. Zhang, T. Suemoto, M. Yoshita and H. Akiyama, Optics Express **22** (2014) 4196-4201.

8. Analysis of Photoexcitation Energy Dependence in the Photoluminescence of Firefly Luciferin: M. Hiyama, H. Akiyama, T. Mochizuki, K. Yamada and N. Koga, Photochem. Photobiol. **90** (2014) 820-828.

---

* Joint research among groups within ISSP.

9. Atomically Engineered Metal-Insulator Transition at the $TiO_2/LaAlO_3$ Heterointerface: M. Minohara, T. Tachikawa, Y. Nakanishi, Y. Hikita, L. F. Kourkoutis, J.-S. Lee, C.-C. Kao, M. Yoshita, H. Akiyama, C. Bell and H. Y. Hwang, Nano Lett. **14 (11)** (2014) 6743-6746.

10. Gain switching of a double-core-waveguide semiconductor laser via traveling-wave optical pumping: H. Nakamae, T. Nakamura, T. Ito, T. Mochizuki, C. Kim, S. Chen, M. Yoshita and H. Akiyama, Appl. Phys. Express **7** (2014) 062701.

11. *Direct generation of 2-ps blue pulses from gain-switched InGaN VCSEL assessed by up-conversion technique: A. Asahara, S. Chen, T. Ito, M. Yoshita, W. Liu, B. Zhang, T. Suemoto and H. Akiyama, Sci. Rep. **4** (2014) 6401.

12. *Spectral dynamics of picosecond gain-switched pulses from nitride-based vertical-cavity surface-emitting lasers: S. Chen, T. Ito, A. Asahara, M. Yoshita, W. Liu, J. Zhang, B. Zhang, T. Suemoto and H. Akiyama, Sci. Rep. **4** (2014) 4325.

13. Balance sheets of energy and carriers and subcell characteristics in a InGaP/GaAs/Ge tandem solar cell: S. Chen, L. Zhu, M. Yoshita, T. Mochizuki, C. Kim, H. Akiyama, M. Imaizumi and Y. Kanemitsu, Photovoltaic Specialists Conference Proceedings **IEEE 40th** (2014) 1780-1783.

14. Conversion efficiency limits and optimized designs for tandem solar cells with realistic sub-cell material quality: L. Zhu, T. Mochizuki, M. Yoshita, S. Chen, S. Sato, C. Kim, H. Akiyama and Y. Kanemitsu, Photovoltaic Specialists Conference Proceedings **IEEE 40th** (2014) 3404-3408.

15. Time-resolved photoluminescence measurements for determining voltage-dependent charge-separation efficiencies of subcells in triple-junction solar cells: D. M. Tex, T. Ihara, H. Akiyama, M. Imaizumi and Y. Kanemitsu, Appl. Phys. Lett. **106** (2015) 013905.

16. 多接合太陽電池の診断・設計と発光絶対値・発光量子効率の評価：秋山 英文 , 応用物理 **84** (2015) 319-325.

17. Multi-junction-solar-cell designs and characterizations based on detailed-balance principle and luminescence yields: H. Akiyama, L. Zhu, M. Yoshita, C. Kim, S. Chen, T. Mochizuki and Y. Kanemitsu, Proc. SPIE **9358** (2015) 93580B1-8.

18. Time-resolved observation of excitonic dynamics under coherent terahertz excitation in GaAs quantum wells: K. Uchida, H. Hirori, T. Aoki, C. Wolpert, K. Tanaka, T. Mochizuki, C. Kim, M. Yoshita, H. Akiyama, L. N. Pfeiffer and K. W. West, Proc. SPIE **9361** (2015) 93611L1-6.

19. Analysis of Oxyluciferin Photoluminescence Pathways in Aqueous Solutions: M. Hiyama, T. Mochizuki, H. Akiyama and N. Koga, Photochem. Photobiol. **91** (2015) 74.

20. *Effect of very high magnetic field on the optical properties of firefly light emitter oxyluciferin: W. Zhou, D. Nakamura, Y. Wang, T. Mochizuki, H. Akiyama and S. Takeyama, Journal of Luminescence **165** (2015) 15.

21. On the importance of cavity-length and heat dissipation in GaN-based vertical-cavity surface-emitting lasers: W. J. Liu, X. L. Hu, L. Y. Ying, S. Q. Chen, J. Y. Zhang, H. Akiyama, Z. P. Cai and B. P. Zhang, Sci. Rep. **5** (2015) 9600.

22. Thorough subcells diagnosis in a multi-junction solar cell via absolute electroluminescence-efficiency measurements: S. Chen, L. Zhu, M. Yoshita, T. Mochizuki, C. Kim, H. Akiyama, M. Imaizumi and Y. Kanemitsu, Sci. Rep. **5** (2015) 7836.

23. *First-Principles Investigation of Strong Excitonic Effects in Oxygen 1s X-ray Absorption Spectra: Y. Noguchi, M. Hiyama, H. Akiyama, Y. Harada and N. Koga, J. Chem. Theory Comput. **11** (2015) 1668-1673.

24. Strong localization effect and carrier relaxation dynamics in self-assembled InGaN quantum dots emitting in the green: G.-E. Weng, W.-R. Zhao, S.-Q. Chen, H. Akiyama, Z.-C. Li, J.-P. Liu and B.-P. Zhang, Nanoscale Res. Lett. **10** (2015) 31.

25. *Vibronic Structures in Absorption and Fluorescence Spectra of Firefly Oxyluciferin in Aqueous Solutions: M. Hiyama, Y. Noguchi, H. Akiyama, K. Yamada and N. Koga, Photochem Photobiol (2015) published online, in print.

I. Matsuda group

Developments and experiments of the advanced spectroscopies have been carried out by using vacuum ultraviolet (VUV) and soft X-rays (SX). At SPring-8 BL07LSU, we have adjusted phase shifters with electromagnetic coils of the segmented cross-type undulator in the electron storage ring to realize fast switching of the light polarizations. At the end-station, we routinely supported picosecond-time-resolved SX photoemission spectroscopy experiments of joint-researches. Studies of photovoltaics and photocatalysis have been carried out mainly. As a new experimental tool for investigating spin dynamics, we have continued development of the rotating analyzer ellipsometry method for measuring the resonant magneto-optical Ker effect (RMOKE) using VUV and SX rays. Using a seeded free-electron laser at FERMI@ELETTRA in Italy, we have succeeded in tracing the photo-induced femtoseconds-spin-flipping in the ferrimagnetic material by RMOKE.

† Joint research with outside partners.

1. Anisotropic electronic conduction in metal nanofilms grown on a one-dimensional surface superstructure: N. Nagamura, R. Hobara, T. Uetake, T. Hirahara, K. Kobayashi, I. Matsuda and S. Hasegawa, Phys. Rev. B **89** (2014) 125415-1,-5.

2. †*Observation of a giant Kerr rotation in a ferromagnetic transition metal by M-edge resonant magneto-optic Kerr effect: Sh. Yamamoto, M. Taguchi, M. Fujisawa, R. Hobara, S. Yamamoto, K. Yaji, T. Nakamura, K. Fujikawa, R. Yukawa, T. Togashi, M. Yabashi, M. Tsunoda, S. Shin and I. Matsuda, Phys. Rev. B **89** (2014) 064423(1-6).

3. Electron-hole recombination on ZnO(0001) single-crystal surface studied by time-resolved soft X-ray photoelectron spectroscopy: R. Yukawa, S. Yamamoto, K. Ozawa, M. Emori, M. Ogawa, Sh. Yamamoto, K. Fujikawa, R. Hobara, S. Kitagawa, H. Daimon, H. Sakama and I. Matsuda, Appl. Phys. Lett. **105** (2014) 151602-1,-4.

4. †*Observing hot carrier distribution in an n-type epitaxial graphene on a SiC substrate: T. Someya, H. Fukidome, Y. Ishida, R. Yoshida, T. Iimori, R. Yukawa, K. Akikubo, Sh. Yamamoto, S. Yamamoto, T. Yamamoto, T. Kanai, K. Funakubo, M. Suemitsu, J. Itatani, F. Komori, S. Shin and I. Matsuda, Appl. Phys. Lett. **104** (2014) 161103(1-4).

5. Non-linear kinetic model for oscillatory relaxation of the photovoltage effect on a Si(111)7x7 surface: M. Ogawa, R. -Y. Liu, C. -H. Lin, S. Yamamoto, R. Yukawa, R. Hobara, S. -J. Tang and I. Matsuda, Surf. Sci. **624** (2014) 70-75.

6. 反射高速陽電子回折（RHEPD）による Ag(111) 表面上のシリセンの構造決定：深谷 有喜，望月 出海，前川 雅樹，和田 健，兵頭 俊夫，松田 巌，河裾 厚男，PF News **32** (2014) 10-14.

7. 表面電子化合物：松田 巌，深谷 有喜，PF News **31** (2014) 33-37.

8. †Electron–Hole Recombination Time at TiO_2 Single-Crystal Surfaces: Influence of Surface Band Bending: K. Ozawa, M. Emori, S. Yamamoto, R. Yukawa, S. Yamamoto, R. Hobara, K. Fujikawa, H. Sakama and I. Matsuda, J. Phys. Chem. Lett. **5** (2014) 1953-1957.

9. Rashba effects within the space charge layer of a semiconductor: C.-H. Lin, T.-R. Chang, Ro. -Ya. Liu, C.-M. Cheng, K.-D. Tsuei, H. -T. Jeng, C.-Y. Mou, I. Matsuda and S. -J. Tang, New. J. Phys. **16** (2014) 045003-1,-12.

10. †*New soft X-ray beamline BL07LSU at SPring-8: S. Yamamoto, Y. Senba, T. Tanaka, H. Ohashi, T. Hirono, H. Kimura, M. Fujisawa, J. Miyawaki, A. Harasawa, T. Seike, S. Takahashi, N. Nariyama, T. Matsushita, M. Takeuchi, T. Ohata, Y. Furukawa, K. Takeshita, S. Goto, Y. Harada, S. Shin, H. Kitamura, A. Kakizaki, M. Oshima and I. Matsuda, J Synchrotron Rad **21** (2014) 352-365.

11. †*Development of an electromagnetic phase shifter using a pair of cut-core coils for a cross undulator: I. Matsuda, A. Kuroda, J. Miyawaki, Y. Kosegawa, S. Yamamoto, T. Seike, T. Bizen, Y. Harada, T. Tanaka and H. Kitamura, Nucl. Instrum. Methods Phys. Res. A **767** (2014) 296-299.

12. †Titanium-induced charge of Si(001) surface dependent on local configuration: K. Shudo, T. Aoki, S. Ohno, K. Yamazaki, F. Nakayama, M. Tanaka, T. Okuda, A. Harasawa, I. Matsuda, T. Kakizaki and M. Uchiyama, J. Elec. Spec. Rel, Phen. **192** (2014) 35.

13. 時間分解軟X線光電子分光法による表面キャリアダイナミクス研究の進展：山本 達，松田 巌，日本放射光学会誌 **27** (2014) 241.

14. *Scanning tunneling spectroscopy study of quasiparticle interference on dual topological insulator $Bi_{1-x}Sb_x$: S. Yoshizawa, F. Nakamura, A. A. Taskin, T. Iimori, K. Nakatsuji, I. Matsuda, Y. Ando and F. Komori, Phys. Rev. B **91** (2015) 045423-1,-6.

15. †Experimental evidence for two-dimensional states localized in subsurface region in Ge(111): K. Yaji, Y. Ohtsubo, S. Hatta, H. Okuyama, R. Yukawa, I. Matsuda, P. Le Fevre, F. Bertran, A. Taleb-Ibrahimi, A. Kakizaki and T. Aruga, J. Elec. Spec. Rel. Phenom. (2014), in print.

Kobayashi group

We have demonstrated high-power high-harmonic generation from 30-m cavity. The obtained VUV coherent light had a comparable brightness to a synchrotron radiation. We are developing very-high repetition-rate mode-locked laser for astronomical applications. Long-term stable optical frequency comb is also developing. We have stated to develop a mid-infrared optical frequency comb for medical applications.

1. *Time-resolved photoemission apparatus achieving sub-20-meV energy resolution and high stability: Y. Ishida, T. Togashi, K. Yamamoto, M. Tanaka, T. Kiss, T. Otsu, Y. Kobayashi and S. Shin, Rev. Sci. Instrum. **85** (2014) 123904(1-9).

2. *Wavefront analysis of high-efficiency, large-scale, thin transmission gratings: C. Zhou, T. Seki, T. Kitamura, Y.

* Joint research among groups within ISSP.

Kuramoto, T. Sukegawa, N. Ishii, T. Kanai, J. Itatani, Y. Kobayashi and S. Watanabe, Opt. Express **22** (2014) 5995-6008.

3. デュアルコム分光 FT-IR にかわる高速広帯域精密分光 : 久世 直也 , 小澤 陽 , 小林 洋平 , 日本物理学会 **69** (2014) 29.

4. エンハンスメントキャビティー、フォトンリングを用いた高強度レーザー実現の試み : 小林 洋平 , プラズマ・核融合学会誌 第 90 巻 8 号 (2014) 462-467.

5. 300-mW narrow-linewidth deep-ultraviolet light generation at 193 nm by frequency mixing between Yb-hybrid and Er-fiber lasers: H. Xuan, Z. Zhao, H. Igarashi, S. Ito, K. Kakizaki and Y. Kobayashi, Opt. Express **23** (2015) 10564-10572.

6. Direct 15-GHz mode-spacing optical frequency comb with a Kerr-lens mode-locked Yb:Y_2O_3 ceramic laser: M. Endo, I. Ito and Y. Kobayashi, Opt. Express **23** (2015) 1276-1282.

7. *Emergent photovoltage on SmB_6 surface upon bulk-gap evolution revealed by pump-and-probe photoemission spectroscopy: Y. Ishida, T. Otsu, T. Shimada, M. Okawa, Y. Kobayashi, F. Iga, Y. Takabatake and S. Shin, Sci. Rep. **5** (2015) 8160(1-6).

8. Development of compact and ultra-high-resolution spectrograph with multi-GHz optical frequency comb: M. Endo, T. Sukegawa, A. Silva and Y. Kobayashi, in: *Ground-based and Airborne Instrumentation for Astronomy V* (Proc. SPIE 9147, 2014), 91477Y.

9. Development of narrow-linewidth Yb- and Er- fiber lasers and frequency mixing for ArF excimer laser seeding: H. Xuan, Z. Zhao, H. Igarashi, S. Ito, K. Kakizaki and Y. Kobayashi, in: *Fiber Lasers XI: Technology, Systems, and Applications* (Proc. SPIE 8961, 2014), 89612M.

Itatani group

We have continued to develop an infrared (IR) optical parametric chirped pulse amplifier (OPCPA) to produce coherent soft X ray via high harmonic generation. Comparison between the observed carrier-envelope phase dependence of high harmonic spectra and a quantum simulation suggested the generation of isolated attosecond pulses that covered the carbon K edge(~284 eV). This is an important milestone for realizing ultrafast soft-X-ray spectroscopy using laser-based light sources. The long-term stability of the carrier-envelope phase of the IR OPCPA was examined by monitoring the high-energy photoelectron spectra in above-threshold ionization, revealing the excellent long-term passive stabilization up to 47 hours. The molecular control experiments using intense THz pulses was extended to a novel double-pulse scheme. We successfully controlled the two-level coherence that were probed by the velocity map technique. We also kept collaboration with Shin, Komori, and Matsuda groups at LASOR-ISSP on time-resolved ARPES using a femtosecond EUV source. Several experiments on the ultrafast relaxation of photo-induced states in VO_2 and graphene were successfully performed.

1. †*Ultrafast photoinduced transition of an insulating VO_2 thin film into a nonrutile metallic state: R. Yoshida, T. Yamamoto, Y. Ishida, H. Nagao, T. Otsuka, K. Saeki, Y. Muraoka, R. Eguchi, K. Ishizaka, T. Kiss, S. Watanabe, T. Kanai, J. Itatani and S. Shin, Phys. Rev. B **89** (2014) 205114(1-7).

2. †*Observing hot carrier distribution in an n-type epitaxial graphene on a SiC substrate: T. Someya, H. Fukidome, Y. Ishida, R. Yoshida, T. Iimori, R. Yukawa, K. Akikubo, Sh. Yamamoto, S. Yamamoto, T. Yamamoto, T. Kanai, K. Funakubo, M. Suemitsu, J. Itatani, F. Komori, S. Shin and I. Matsuda, Appl. Phys. Lett. **104** (2014) 161103(1-4).

3. 高次高調波によるコヒーレント軟 X 線の発生とその応用 : 板谷 治郎 , 光学 **43** (2014) 419-425.

4. Selecting rotational two-level coherence in polar molecules by double terahertz pulses: K. Kitano, N. Ishii, T. Kanai and J. Itatani, Phys. Rev. A **90** (2014) 041402(R)-1-5.

5. Long-term passive stabilization of the carrier-envelope phase of an intense infrared few-cycle pulse source: H. Geiseler, N. Ishii, K. Kaneshima, T. Kanai and J. Itatani, Appl. Phys. B **117** (2014) 941-946.

6. High-energy half-cycle cutoffs in high harmonic and rescattered electron spectra using waveform-controlled few-cycle infrared pulses: H. Geiseler, N. Ishii, K. Kaneshima, K. Kitano, T. Kanai and J. Itatani, J. Phys. B: At. Mol. Opt. Phys. **47** (2014) 204011-1-8.

7. *Wavefront analysis of high-efficiency, large-scale, thin transmission gratings: C. Zhou, T. Seki, T. Kitamura, Y. Kuramoto, T. Sukegawa, N. Ishii, T. Kanai, J. Itatani, Y. Kobayashi and S. Watanabe, Opt. Express **22** (2014) 5995-6008.

8. Carrier-envelope phase-dependent high harmonic generation in the water window using few-cycle infrared pulses: N. Ishii, K. Kaneshima, K. Kitano, T. Kanai, S. Watanabe and J. Itatani, Nat. Commun. **5** (2014) 3331.

9. Ultrabroadband infrared chirped mirrors characterized by a white-light Michelson interferometer: K. Kaneshima, M.

† Joint research with outside partners.

Sugiura, K. Tamura, N. Ishii and J. Itatani, Appl. Phys. B **119** (2015) 347-353.

Harada group

Soft X-ray absorption/emission spectroscopy in a real ambient condition is an urgent requisite since the ultrahigh vacuum needed to transmit soft X-rays hampers the use of this unique method among various spectroscopies for a long time. Therefore, this is one of the major goals of our project. From 2013, we have developed a system for ambient pressure soft X-ray emission spectroscopy using originally-developed differential pumping system. In 2014, we have introduced a co-axial sample monitor and a beam profile monitor, which has facilitated the alignment of the apertures and a nozzle for differential pumping to efficiently transmit incident soft X-rays to the sample. Finally we have enabled 10 orders of magnitude different pressure (from 10^{-5} Pa to 10^{5} Pa) at only around 400 mm distance. As a demonstration we performed O 1s RIXS of H_2O ice under the pressure around 10^{-1}Pa. We also have introduced a system to apply a magnetic field on the sample up to 0.3 T. We have accepted 7 collaborative works at BL07LSU HORNET endstation, which are i) Observation of 100 meV-order multiphonon excitations in Ti 2p RIXS of anatase TiO_2, which reveals real bulk sensitive isotropic phonon modes for the first time; ii) Development of damage-free RIXS technique for highly valent manganese complex as a model manganese cluster for oxygen evolving complex in photosystem II; iii) Study on the structure making/breaking characteristics of hydrated water around a series of hydrophobic phase-transfer catalysts: TBA, TPA, TEA and TMA, which followed a general trend even in the electronic structure level from structure-breaking to structure-making as the size of the catalyst decreases; iv) MCD-RIXS experiments applying the 0.3 T magnetic field, one is terfenol-D, the most famous magnetostrictive material and the other is half metal CrO_2 which demonstrated a beautiful selection rule of dd excitations depending on the polarization of the incident photon due to fully spin-polarized HOMO state under the magnetic field; v) Operando Fe 2p RIXS spectroscopy of the cathode electrode $LiFePO_4$ for lithium ion batteries by characterizing electrochemical property.

1. †Unveiling the impurity band induced ferromagnetism in the magnetic semiconductor (Ga,Mn)As: M. Kobayashi, I. Muneta, Y. Takeda, Y. Harada, A. Fujimori, J. Krempaský, T. Schmitt, S. Ohya, M. Tanaka, M. Oshima and V. N. Strocov, Phys. Rev. B **89** (2014) 205204(1-8).

2. †Electronic Excitations of a Magnetic Impurity State in the Diluted Magnetic Semiconductor (Ga,Mn)As: M. Kobayashi, H. Niwa, Y. Takeda, A. Fujimori, Y. Senba, H. Ohashi, A. Tanaka, S. Ohya, P. N. Hai, M. Tanaka, Y. Harada and M. Oshima, Phys. Rev. Lett. **112** (2014) 107203(1-7).

3. Reabsorption of Soft X-Ray Emission at High X-Ray Free-Electron Laser Fluences: S. Schreck, M. Beye, J. A. Sellberg, T. McQueen, H. Laksmono, B. Kennedy, S. Eckert, D. Schlesinger, D. Nordlund, H. Ogasawara, R. G. Sierra, V. H. Segtnan, K. Kubicek, W. F. Schlotter, G. L. Dakovski, S. P. Moeller, U. Bergmann, S. Techert, L. G. M. Pettersson, P. Wernet, M. J. Bogan, Y. Harada, A. Nilsson and A. Föhlisch, Phys. Rev. Lett. **113** (2014) 153002(1-6).

4. Spin and orbital magnetic moments of Fe in the n-type ferromagnetic semiconductor (In,Fe)As: M. Kobayashi, L. D. Anh, P. N. Hai, Y. Takeda, S. Sakamoto, T. Kadono, T. Okane, Y. Saitoh, H. Yamagami, Y. Harada, M. Oshima, M. Tanaka and A. Fujimori, Appl. Phys. Lett. **105** (2014) 032403(1-4).

5. †*Solvation dependence of valence electronic states of water diluted in organic solvents probed by soft X-ray spectroscopy: T. Tokushima, Y. Horikawa, O. Takahashi, H. Arai, K. Sadakane, Y. Harada, Y. Takata and S. Shin, Phys. Chem. Chem. Phys. **16** (2014) 10753-10761.

6. †Distinguishing between High- and Low-Spin States for Divalent Mn in Mn-Based Prussian Blue Analogue by High-Resolution Soft X-ray Emission Spectroscopy: D. Asakura, Y. Nanba, M. Okubo, Y. Mizuno, H. Niwa, M. Oshima, H. Zhou, K. Okada and Y. Harada, J. Phys. Chem. Lett. **5** (2014) 4008-4013.

7. †*New soft X-ray beamline BL07LSU at SPring-8: S. Yamamoto, Y. Senba, T. Tanaka, H. Ohashi, T. Hirono, H. Kimura, M. Fujisawa, J. Miyawaki, A. Harasawa, T. Seike, S. Takahashi, N. Nariyama, T. Matsushita, M. Takeuchi, T. Ohata, Y. Furukawa, K. Takeshita, S. Goto, Y. Harada, S. Shin, H. Kitamura, A. Kakizaki, M. Oshima and I. Matsuda, J Synchrotron Rad **21** (2014) 352-365.

8. Iron-Nitrogen Coordination in Modified Graphene Catalyzes a Four-Electron-Transfer Oxygen Reduction Reaction: K. Kamiya, H. Koshikawa, H. Kiuchi, Y. Harada, M. Oshima, K. Hashimoto and S. Nakanishi, ChemElectroChem **1** (2014) 877-884.

9. †*Development of an electromagnetic phase shifter using a pair of cut-core coils for a cross undulator: I. Matsuda, A. Kuroda, J. Miyawaki, Y. Kosegawa, S. Yamamoto, T. Seike, T. Bizen, Y. Harada, T. Tanaka and H. Kitamura, Nucl. Instrum. Methods Phys. Res. A **767** (2014) 296-299.

10. †Multi-Phonon Excitations in Fe 2*p* RIXS on Mg_2FeH_6: K. Kurita, D. Sekiba, I. Harayama, K. Chito, Y. Harada, H. Kiuchi, M. Oshima, S. Takagi, M. Matsuo, R. Sato, K. Aoki and S.-I. Orimo, J. Phys. Soc. Jpn. **84** (2015) 043201 1-3.

11. X-ray Emission Spectroscopy of Bulk Liquid Water in "No-man's Land": J. Sellberg, T. McQueen, H. Laksmono, S.

* Joint research among groups within ISSP.

Schreck, M. Beye, D. DePonte, B. O'Kennedy, D. Nordlund, R. Sierra, D. Schlesinger, T. Tokushima, S. Eckert, V. Segtnan, H. Ogasawara, K. Kubicek, S. Techert, U. Bergmann, G. Dakovski, W. Schlotter, Y. Harada, I. Zhovtobriukh, M. Bogan, P. Wernet, A. Föhlisch, L. Pettersson and A. Nilsson, J. Chem. Phys. **142** (2015) 044505 1-9.

12. †Enhancement in Kinetics of the Oxygen Reduction Reaction on a Nitrogen-Doped Carbon Catalyst by Introduction of Iron via Electrochemical Methods: J. Wu, D. Zhang, H. Niwa, Y. Harada, M. Oshima, H. Ofuchi, Y. Nabae, T. Okajima and T. Ohsaka, Langmuir **31** (2015) 5529-5536.

13. Active site formation mechanism of carbon-based oxygen reduction catalysts derived from a hyperbranched iron phthalocyanine polymer: Y. Hiraike, M. Saito, H. Niwa, M. Kobayashi, Y. Harada, M. Oshima, J. Kim, Y. Nabae and M.-A. Kakimoto, Nanoscale Res Lett **10** (2015) 179 1-11.

14. †Operando soft x-ray emission spectroscopy of $LiMn_2O_4$ thin film involving Li-ion extraction/insertion reaction: D. Asakura, E. Hosono, H. Niwa, H. Kiuchi, J. Miyawaki, Y. Nanba, M. Okubo, H. Matsuda, H. S. Zhou, M. Oshima and Y. Harada, Electrochem. Commun. **50** (2015) 93-96.

15. *First-Principles Investigation of Strong Excitonic Effects in Oxygen 1s X-ray Absorption Spectra: Y. Noguchi, M. Hiyama, H. Akiyama, Y. Harada and N. Koga, J. Chem. Theory Comput. **11** (2015) 1668-1673.

16. カーボンアロイ触媒の開発、そしてその可能性：難波江 裕太 , 原田 慈久 , 尾嶋 正治 , (シーエムシー出版 , SPring-8 の高輝度放射光を利用したグリーンエネルギー分野における電池材料開発、第５章電池における触媒開発 , 2014).

17. * 鉄タンパク質の電子状態と機能：原田 慈久 , 辛 埴 , (朝倉書店「鉄の事典」, 3.11.d 節 , 2014).

Wadati group

Our main experimental techniques are synchrotron-based x-ray spectroscopy and scattering. We studied the orbital and magnetic phase transitions in $Pr_{0.5}Ca_{0.5}MnO_3/La_{Sr}Ca_{0.5}MnO_3$ superlattices by resonant soft x-ray scattering and observed dimensional crossover as a function of temperature. We also performed time-resolved x-ray diffraction study of $Pr_{0.5}Ca_{0.5}MnO_3$ thin films by using x-ray free electron laser in LCLS (USA), and observed a time-dependent order parameter by creating coherent phonons.

1. Antiferromagnetic Order of the Co^{2+} High-Spin State with a Large Orbital Angular Momentum in $La_{1.5}Ca_{0.5}CoO_4$: J. Okamoto, H. Nakao, Y. Yamasaki, H. Wadati, A. Tanaka, M. Kubota, K. Horigane, Y. Murakami and K. Yamada, J. Phys. Soc. Jpn. **83** (2014) 044705(1-6).

2. Insulator-to-Superconductor Transition upon Electron Doping in a BiS_2-Based Superconductor $Sr_{1-x}La_xFBiS_2$: H. Sakai, D. Kotajima, K. Saito, H. Wadati, Y. Wakisaka, M. Mizumaki, K. Nitta, Y. Tokura and S. Ishiwata, J. Phys. Soc. Jpn. **83** (2014) 014709(1-7).

3. Bond order and the role of ligand states in stripe-modulated $IrTe_2$: K. Takubo, R. Comin, D. Ootsuki, T. Mizokawa, H. Wadati, Y. Takahashi, G. Shibata, A. Fujimori, R. Sutarto, F. He, S. Pyon, K. Kudo, M. Nohara, G. Levy, I. Elfimov, G. A. Sawatzky and A. Damascelli, Phys. Rev. B **90** (2014) 081104(R)(1-5).

4. Photoemission and DMFT study of electronic correlations in $SrMoO_3$: Effects of Hund's rule coupling and possible plasmonic sideband: H. Wadati, J. Mravlje, K. Yoshimatsu, H. Kumigashira, M. Oshima, T. Sugiyama, E. Ikenaga, A. Fujimori, A. Georges, A. Radetinac, K. S. Takahashi, M. Kawasaki and Y. Tokura, Phys. Rev. B **90** (2014) 205131(1-8).

5. Orbital correlations and dimensional crossover in epitaxial $Pr_{0.5}Ca_{0.5}MnO_3/La_{0.5}Sr_{0.5}MnO_3$ superlattices: H. Wadati, J. Okamoto, M. Garganourakis, V. Scagnoli, U. Staub, E. Sakai, H. Kumigashira, T. Sugiyama, E. Ikenaga, M. Nakamura, M. Kawasaki and Y. Tokura, New J. Phys. **16** (2014) 073044(1-10).

6. Revealing orbital and magnetic phase transitions in $Pr_{0.5}Ca_{0.5}MnO_3$ epitaxial thin films by resonant soft x-ray scattering: H. Wadati, J. Geck, E. Schierle, R. Sutarto, F. He, D. G. Hawthorn, M. Nakamura, M. Kawasaki, Y. Tokura and G. A. Sawatzky, New J. Phys. **16** (2014) 033006(1-12).

7. A time-dependent order parameter for non-equilibrium phase transitions: P. Beaud, A. Caviezel, S. O. Mariager, L. Rettig, G. Ingold, C. Dornes, S.-W. Huang, J. A. Johnson, M. Radovic, T. Huber, T. Kubacka, A. Ferrer, H. T. Lemke, M. Chollet, D. Zhu, J. M. Glownia, M. Sikorski, A. Robert, H. Wadati, M. Nakamura, M. Kawasaki, Y. Tokura, S. L. Johnson and U. Staub, Nat. Mater. **13** (2014) 923-927.

8. X-ray spectroscopic study of $BaFeO_3$ thin films; an Fe^{4+} ferromagnetic insulator: T. Tsuyama, T. Matsuda, S. Chakraverty, J. Okamoto, E. Ikenaga, A. Tanaka, T. Mizokawa, H. Y. Hwang, Y. Tokura and H. Wadati, Phys. Rev. B **91** (2015) 115101(1-7).

9. Engineering a spin-orbital magnetic insulator by tailoring superlattices: J. Matsuno, K. Ihara, S. Yamamura, H. Wadati, K. Ishii, V. V. Shankar, H.-Y. Kee and H. Takagi, Phys. Rev. Lett. **114** (2015) 247209(1-5).

† Joint research with outside partners.

10. Observation of a Devil's Staircase in the Novel Spin-Valve System $SrCo_6O_{11}$: T. Matsuda, S. Partzsch, T. Tsuyama, E. Schierle, E. Weschke, J. Geck, T. Saito, S. Ishiwata, Y. Tokura and H. Wadati, Phys. Rev. Lett. **114** (2015) 236403(1-5).

Kondo group

A central issue in the search for the mechanism of high temperature conductivity is to determine the bosons which mediate electron pairing. Electron coupling with a bosonic mode changes the slope of the band dispersion within its excitation energy. Such a band renormalization or ''kink''is usually observed in superconducting materials. By using laser-based angle-resolved photoemission spectroscopy, capable of ultrahigh energy resolution, we found a universal relation between electron-mode coupling and Tc in the pnictide high-Tc superconductor.

1. †*Evidence of a universal relation between electron-mode coupling and T_c in $Ba_{1-x}K_xFe_2As_2$ superconductor from laser angle-resolved photoemission spectroscopy: W. Malaeb, T. Shimojima, Y. Ishida, T. Kondo, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, T. Saito, Y. Kohori and S. Shin, Phys. Rev. B **90** (2014) 195124(1-6).

2. Fermi Surface and Pseudogap Evolution in a Cuprate Superconductor.: Y. He, Y. Yin, M. Zech, A. Soumyanarayanan, M. M. Yee, T. Williams, M. C. Boyer, K. Chatterjee, W. D. Wise, I. Zeljkovic, T. Kondo, T. Takeuchi, H. Ikuta, P. Mistark, R. S. Markiewicz, A. Bansil, S. Sachdev, E. W. Hudson and J. E. Hoffman, Science **344** (2014) 608-611.

3. Pairing, pseudogap and Fermi arcs in cuprates.: A. Kaminski, T. Kondo, T. Takeuchi and G. Gu, Philosophical Magazine **95** (2015) 453-466.

Okazaki group

We have developed and improved a time- and angle-resolved photoemission (TARPES) apparatus using an EUV and SX laser by high harmonics generation (HHG). In 2014, we have installed a new chamber for HHG with a gas cell, a spectrometer for checking spectra of high harmonics, and a chamber for multi-layer mirrors to select a single order of the harmonics. Now, we can use 28, 40, and 70 eV for a probe light. We have also installed an optical parametric amplifier (OPA) system for a pump light, and succeeded in obtaining TRPES spectra of graphite using this system. We have studied nonequilibrium electronic states of graphenes, iron-based superconductors, strongly correlated electron systems, and valence fluctuating heavy fermion systems. In addition, we have also investigated superconducting-gap structures of iron-based superconductors and BiS2-based superconductors by a low-temperature and high-resolution laser ARPES apparatus.

1. *Evidence for excluding the possibility of d-wave superconducting-gap symmetry in Ba-doped KFe_2As_2: Y. Ota, K. Okazaki, Y. Kotani, T. Shimojima, W. Malaeb, S. Watanabe, C. -T. Chen, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, T. Saito, H. Fukazawa, Y. Kohori and S. Shin, Phys. Rev. B **89** (2014) 081103(1-5).

2. †*Evidence of a universal relation between electron-mode coupling and T_c in $Ba_{1-x}K_xFe_2As_2$ superconductor from laser angle-resolved photoemission spectroscopy: W. Malaeb, T. Shimojima, Y. Ishida, T. Kondo, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, T. Saito, Y. Kohori and S. Shin, Phys. Rev. B **90** (2014) 195124(1-6).

3. *Superconductivity in an electron band just above the Fermi level: possible route to BCS-BEC superconductivity: K. Okazaki1, Y. Ito, Y. Ota, Y. Kotani, T. Shimojima, T. Kiss, S. Watanabe, C. -T. Chen, S. Niitaka, T. Hanaguri, H. Takagi, A. Chainani and S. Shin, Sci. Rep. **4** (2014) 4109(1-6).

* Joint research among groups within ISSP.