Division of Condensed Matter 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 2015 include: (1) Discovery of a magnetic field-induced phase transition in PrTi_2Al_2O, a f-electron system with the Gamma-3 non-magnetic doublet crystal field ground state, by means of Al-NMR and magnetization measurements. (2) First observation of a magnetic order in the heavy electron system beta-YbAlB_4 under high pressure of 8 GPa.


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 2016. (1) An experimental technique of low-temperature magnetization measurements under in-situ tuning of the sample orientation has been developed. A two-axis goniometer is introduced into our capacitively-detected Faraday magnetometer, making it possible to rotate the sample by |θ|<7 deg. and |φ|<3 deg. The apparatus has been applied to a metamagnetic heavy fermion compound CeRu_2Si_2, and found that the magnetization isotherms $M(H)$ of this tetragonal compound can be completely scaled by $H \cos \theta$, where $\theta$ is the angle between $H$ and the $c$ axis, confirming the Ising nature of this system. (2) The critical exponent ν of the phase boundary of a field-induced incommensurate ordering state has been examined on the spin-1/2 ferromagnetic-leg ladder 3-Br-4-F-V [=3-(3-bromo-4-fluorophenyl)-1,5-diphenylverdazyl]. Using the temperature-window fitting technique, we obtained the critical exponents which agreed with the 3D Bose-Einstein condensation (BEC) universality class at both sides of the lower critical field and the saturation field. 3-Br-4-F-V thus becomes a new member of the quantum magnets which prove the universality of the 3D BEC exponent. (3) Quasiparticle excitations in the hexagonal heavy-fermion superconductor UPd_2Al_3 have been studied by means of heat capacity ($C$) measurements under rotating magnetic fields. At low temperatures, the polar angle ($\theta$) dependence of $C$ exhibits a maximum along $H||[0001]$ with a two-fold symmetric oscillation below 0.5 T, and an unusual shoulder/hump anomaly has been found around 30-60 deg. from the $c$ axis in $C(\theta)$ at intermediate fields (1<$H$<2 T). This feature in $C(\theta)$ entirely comes from nodal quasiparticle excitations, and has been successfully explained by theoretical calculations assuming a gap function with a horizontal line node.


* Joint research among groups within ISSP.


† Joint research with outside partners.


Mori group

We have successfully developed and unveiled unprecedented functional properties for the molecular materials. The major achievements in 2016 are (1) to disclose the chemical pressure effect by anion substitution on hydrogen-bond-mediated phase transition of \( \beta'\)-H\(_3\)(Cat-EDO-TTF)\(_2\)BF\(_4\), (2) to improve the stability of a metallic state in benzothienobenzothiophene-based molecular conductor, \( \beta\)-[BTBT(OH)\(_2\)]ClO\(_4\), by an effective increase of dimensionality with hydrogen bonds, and (3) to investigate theoretically the H/D isotope effects on phase transition of hydrogen-bonded organic conductor \( \kappa\)-D\(_3\)(Cat-EDT-X)\(_2\) (X = TTF and ST).


Osada group

We have studied the transport properties of the high-mobility thin-film black phosphorus (BP) field effect transistor (FET) device, and have newly found double carrier transport features in positively gated (electron-doped) region. We built a van der Waals stacking structure, hexagonal born nitride (h-BN)/thin-film PB (15-20 nm)/h-BN on SiO\(_2\)/n+-Si substrate, in the groove box environment and then fabricated it into an FET device. By avoiding the degradation of BP by oxygen and water using the h-BN sandwiched structure, the carrier mobility in the FET has reached 6,000 cm\(^2\)/Vs for holes and 5,800 cm\(^2\)/Vs for electrons, which are the highest mobility ever reported. We have newly observed double period Shubnikov-de Haas oscillations, which indicates two Fermi surfaces (FSs), in the highly electron doped region (positively gated region). The second FS is considered to originate from the second electron subband in the ultra-thin BP films.

Yamashita group

We have been studying (1) quantum criticality in heavy-fermion materials by ultra-low temperature cryostat, (2) thermal-Hall conductivity of exotic excitations in frustrated magnets and (3) a new technique for the study of strongly-correlated electron systems. In this year, we have performed (1) Co and In NMR measurements of CeCoIn$_5$ at ultra-low temperatures, (2) thermal Hall measurements in spin liquid materials Ba$_3$CuSb$_2$O$_9$ and alpha-RuCl$_3$ and (3) scanning-Hall measurements of Mn$_3$Sn.


Division of Condensed Matter Theory

Tsunetsugu group

We have completed the study of optical conductivity near antiferromagnetic transition. The main result is the importance of vertex corrections, and this is different from the case of Mott transition, where the vertex corrections have little effects. We also completed the study of quadrupole antiferro orders in a heavy fermion system, and determined the detailed phase diagram in temperature and magnetic field space. Spin singlet order in breathing pyrochlores has been also investigated in detail, and we have found that the spin size essentially change a ground state order. For S>1/2, tetramer orders are stabilized, which contrasts to dimer orders in the S=1/2 case. For this problem, we have developed a systematic scheme of degenerate perturbation tailored for breathing pyrochlores, and derived an effective Hamiltonian for low-energy dynamics.


Kato group

The main research subject of our laboratory is theory of non-equilibrium properties in nanoscale devices. We have studied (1) adiabatic pumping via a quantum dot with Coulomb interaction, (2) current fluctuations in a Kondo-correlated quantum dot, and (3) Andreev bound states in a multi-terminal superconducting device. We have also collaborated with experimentalists in ISSP, and have supported theoretical aspects on (4) pressure dependence of the magnetic ground states in MnP and (5) proximity effect at a single atomic step by STM.


† Joint research with outside partners.


Division of Nanoscale Science

Katsumoto group

We have continued the study of the proximity superconductivity in a ferromagnetic semiconductor (In,Fe)As. The hysteretic behavior of the critical current indicates that the spin-triplet type pairing is realized inside (In,Fe)As. On the other hand, a Heusler ferromagnet Co2FeSi does not show any proximity superconductivity probably due to the clean ferromagnet-superconductor interface. Microwave response of two-dimensional electrons in quantum Hall state has been studied. By applying gate voltage and controlling the effective sample edge, previously-found absorption peaks are greatly enhanced, which result manifest that the peaks correspond to the edge plasmon absorption.


Otani group

This year we tried to expand our research activities toward spintronics utilizing more complex materials in collaboration with the groups working on strongly correlated materials. For example, we have demonstrated magneto-chiral nonreciprocity of volume spin wave propagation in chiral-lattice ferromagnets and spin/charge interconversion using the surface state of topological insulator. We also examined systematically conventional spin Hall effects in Pt as a function of conductivity and found that the spin Hall effect of Pt is tunable. Apart from those, we newly found metal/Bi oxide interface provides an effective spin momentum locking behavior which can be applied for detection of spin currents.


* Joint research among groups within ISSP.

Komori group

Growth of hetero-epitaxial fcc Fe thin films is controlled by using a nanoscale strain-relief mechanism due to the substrate step edges. The lattice expansion/compression caused by the steps is directly observed in atomic-scale STM images accompanied with the local change of the surface electronic states. Ammonia synthesis reaction by atomic hydrogen on the N-adsorbed Cu surfaces was studied using STM. Presence of the clean Cu surface largely enhances the reaction probability owing to the hydrogen diffusion on the surface. Spin-direction of photoelectrons from spin-helical surface states is coherently controlled by linearly-polarized excitation laser light for the Bi2Se3, Bi(0001) and Bi/Ag(111) surfaces. The direction is three-dimensionally detected by a newly developed SARPES.


† Joint research with outside partners.


* Joint research among groups within ISSP.


**Hasegawa group**

Using scanning tunneling microscopy (STM) we investigated transport properties of a monolayer metallic thin film formed on a semiconductor substrate using superconducting pair correlation as a probing signal. The pair correlation was induced onto the two-dimensional (2D) metallic layer by locating superconducting materials on it. Tunneling spectra taken around the proximity area provides information on how the pair correlation spread and decay from the super/normal interface and how step structures on the layer disturbs the spreading. Since these are closely related with electrical conductance, conductivity through the monoatomic layer and a atomic-high step can be estimated through the spatially resolved spectral measurements. We have taken tunneling spectra at 2.1 K around superconducting Pb island structures formed on Pb-induced striped incommensurate phase; a 2D metallic layer formed on Si(111) substrate, and obtained the conductivity through the 2D layer and across a single atomic-height step. It turned out that the steps have a significant contribution to the total resistance even on a nominally flat surface, and that macroscopic monolayer conductance, which includes the step conductance, is quite sensitive to a miscut angle of the substrate. We also demonstrate that a method of compressed sensing is quite effective to improve the quality of obtained results and to save measurement time in the observation of quasi-particle interference (QPI) by STM. Since QPI provides momentum-space information on electronic states of samples, it is a very powerful tool to investigate the electronic states at very low temperature and/or under magnetic field, where photoemission spectroscopy cannot be performed. The QPI measurement is, however, quite time-consuming; it takes a few days to a week for a single dataset. Utilizing the sparseness of the QPI signals in momentum space, we performed a sparsity-inducing algorithm called least absolute shrinkage and selection operator (LASSO), and demonstrated that LASSO recovers a circular QPI pattern of the Ag(111) surface from a dataset whose size is less than that necessary for the conventional Fourier transformation method. Our results demonstrate that the compressed sensing based on the sparse modeling works well in the QPI analysis and that the concept and the procedure should be applied to various subjects in condensed matter physics.


**Lippmaa group**

Our recent work is related to the materials analysis and structural design of photoelectrode materials for photoelectrochemical water splitting reactions. We have studied the process of optically excited d-state relaxation in oxides and discovered a relaxation path related magnetic order in MnO. This work may open a path to control the relaxation rate of excited d-states by external magnetic fields. Another topic is the design of self-organized nanostructures for constructing robust electrodes in oxide thin films to extract photogenerated carriers from a low-mobility oxide semiconductor. Rapid photocarrier recombination limits the maximum achievable efficiency of photoelectrochemical reactions on photoelectrode surfaces because carriers generated

† Joint research with outside partners.
deep in the semiconductor cannot be transported to the surface without trapping and recombination. Embedding metallic nanowires in an oxide matrix helps to reduce the necessary diffusion path length for excited carriers and thus improve the efficiency of photo-induced electrochemical reactions. We have demonstrated the growth of Ir metal nanopillars in SrTiO$_3$ and showed that tubular Schottky junctions form around the nanopillars. Photocarriers generated in the Schottky depletion regions are quickly separated and transported to the film surface through the metallic nanopillars.


**Functional Materials Group**

**Yoshinobu group**

We conducted several research projects in the fiscal year 2016. (1) The activation and hydrogenation of CO$_2$ on clean and Zn-deposited Cu(111) and Cu(997) surfaces studied by AP-XPS, IRAS, and TPD. (2) The surface chemistry of formic acid on clean and Zn-deposited Cu(111) and Cu(997) surfaces studied by SR-PES, IRAS and TPD. (3) Spectroscopic characterization of H-Cu(111), Zn-Cu(111), Pd-Cu and Pd-Ag surfaces by XPS. (4) Spectroscopic characterization of Pt atoms and clusters on graphene using SR-XPS (5) LT-STM study of CO$_2$ on Cu(997) (6) Independently driven four-probe conductivity measurement of organic thin films including pentacene etc. (7) Gas-exposure effects on single organic layer FET


* Joint research among groups within ISSP.


Akiyama group

In 2016, we improved accuracy of absolute electroluminescence-efficiency measurements for multi-junction solar cells by developing an integration-sphere total-flux characterization system. We started time-resolved photo-emission spectroscopy of solar cells. We studied pico- and femto-second short-pulse generation via gain switching in semiconductor lasers via optical pumping and current injection. We studied photo-cleavage and damages of caged luciferins. We made intensive studies on theoretical quantum-chemistry and molecular-dynamics calculations on oxyluciferins and related molecule systems.


† Joint research with outside partners.


12. Theoretical insights into the effect of pH values on oxidation processes in the emission of firefly luciferin in aqueous solution: M. Hiyama, H. Akiyama and N. Koga, Luminescence (2017), accepted for publication.


Sugino group

We have done first-principles study of materials. On the basis of the many-body Green's function method for excited states, we have developed a method for classifying excitons in a molecule which is shown particularly important for designing luminescence materials. We have also developed a wave function theory on the basis of a tensor decomposition method and, as a first step, we successfully approached a magnetic single-impurity problem. Using the density functional theory, we have studied topological materials, magnetism under high pressure, and bioluminescence.


* Joint research among groups within ISSP.
Quantum Materials Group

Oshikawa group

We studied a wide range of problems in quantum condensed matter theory. In particular, we have proposed a new possibility of realizing and designing Kitaev spin liquid in materials, using Metal-Organic Frameworks. Kitaev discovered a remarkable exactly solvable spin model, which has an exotic spin liquid state as the ground state. With its bond-dependent Ising interaction, initially it was regarded as just a theoretical toy model. This perception was changed by Jackeli and Khaliullin, who found that such interactions can naturally arise from spin-orbit interaction in iridates. This led to a flurry of attempts of experimental realization of Kitaev spin liquid in iridates and RbCl$_3$. However, unfortunately, in these materials a sizable Heisenberg antiferromagnetic interaction arises from the direct exchange, resulting in a non-spin liquid ground state. We proposed a novel strategy of using Metal-Organic Frameworks to realize Kitaev spin liquid, and gave supporting theoretical evidences. While it is also based on Jackeli-Khaliullin mechanism, it has advantages in a natural suppression of direct exchanges and in its flexibility and tunability. In particular, it can lead to realization of a variety of three-dimensional Kitaev spin liquids.


Nakatsuji group

Our group explores novel quantum phases and phase transitions in rare-earth and transition metal based compounds. The...

† Joint research with outside partners.
followings are some relevant results obtained in 2016. (1) We discovered second example of an antiferromagnet that exhibits the anomalous Hall effect at room temperature, the chiral antiferromagnet Mn₄Ge. (2) The quantum criticality at ambient pressure in β-YbAlB₄ can be transformed into a magnetic state whose transition temperature reaches a record high value exceeding 30 K. (3) Our thermodynamic and muSR studies have revealed highly frustrated magnetism with significant quantum fluctuations in the Heisenberg spinel antiferromagnet CdYb₂S₄ (4) Our neutron diffraction experiments in collaboration has revealed that disorder in the pyrochlore lattice plays an important role to enhance quantum fluctuations to stabilize a quantum Coulombic phase formed in a quantum spin ice candidate Pr₂Zr₂O₇.


* Joint research among groups within ISSP.


34. 強相関電子系の物質開発:序説 : 中辻 知 , 「物性科学ハン ドブ ッ ク- 概念・現象・物質」, 13, 家 泰弘 , 高田康民 , ( 朝倉書店 , 2016), 929-931.


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**Materials Design and Characterization Laboratory**

**Hiroi group**

The ferromagnetic and conducting magnetic domain walls in the all-in/all-out order of Cd$_2$Os$_2$O$_7$ are studied. The superconductivity and multipolar phase transitions of the pyrochlore oxide Cd$_2$Re$_2$O$_7$ are revisited. Many frustrated spin systems are studied.

[CdCu$_3$(OH)$_6$(NO$_3$)$_2$·H$_2$O] is a structurally perfect kagome antiferromagnet crystallizing in the kapellasite-type. An

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† Joint research with outside partners.
antiferromagnetic order accompanied by a small spontaneous magnetization that surprisingly is confined in the kagome plane sets in at TN ~ 4 K, well below the nearest-neighbor exchange interaction J/kB = 45 K. This suggests that a unique “q = 0” type 120° spin structure with “negative” (downward) vector chirality, which breaks the underlying threefold rotational symmetry of the kagome lattice and thus allows a spin canting within the plane, is exceptionally realized in this compound rather than a common one with “positive” (upward) vector chirality. The origin is discussed in terms of the Dzyaloshinskii-Moriya interaction. The quasi-1D antiferromagnet NaCuMoO4(OH), which comprises edge-sharing CuO2 chains, is shown to be a good candidate for the frustrated J1–J2 chain model with J1 = 51 K and J2 = 36 K. We are now looking for evidence of a spin nematic state expected just below the saturation field of 26 T. AMoOPO4Cl (A = K, Rb) with Mo5+ ions in the 4d1 electronic configuration are good model compounds for the spin-1/2 J1–J2 square-lattice magnet. Magnetic transitions are observed at around 6 and 8 K in the K and Rb compounds, respectively. In contrast to the normal Néel-type antiferromagnetic order, the NMR and neutron diffraction experiments find a columnar antiferromagnetic order for each compound, which is stabilized by a dominant antiferromagnetic J2. Both compounds realize the unusual case of two interpenetrating J2 square lattices weakly coupled to each other by J1.

1. Pressure-induced non-superconducting phase of β-Na0.33V2O5 and the mechanism of high-pressure phase transitions in β-Na0.33V2O5 and β-Li0.33V2O5 at room temperature: A. Grzechnik, Y. Ueda, T. Yamauchi, M. Hanfland, P. Hering, V. Potapkin and K. Friese, J. Phys.: Condens. Matter 28 (2016) 035401(1-9).


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 activities of 2016 include: (1) development of the tensor network algorithms and codes suitable for parallel computation, (2) application of the tensor network methods to frustrated spin / fermion systems, and (3) quantum Monte Carlo simulation of bosonic systems targeting the two-dimensional Helium system.


Uwatoko group

Temperature dependence of resistivity on KFe$_2$As$_2$ single crystals down to 20 mK was measured under various hydrostatic pressures up to 17.5 GPa generated in a cubic-anvil cell. With increasing the pressure, the superconducting transition of tetragonal KFe$_2$As$_2$ was suppressed gradually and disappears completely at ~ 11 GPa, which was related to the weakening of electronic correlations and/or critical fluctuations under pressure. MnP, a superconductor under pressure, exhibits a ferromagnetic order below $T_C$ ~ 290 K followed by a helical order with the spins lying in the ab plane and the helical rotation propagating along the c axis below $T_s$ ~ 50 K at ambient pressure. Both $T_C$ and $T_s$ are gradually suppressed with increasing pressure and the helical order disappears at ~ 1.2 GPa. At intermediate pressures of 1.8 and 2.0 GPa, the ferromagnetic order first develops and changes to a conical or two-phase (ferromagnetic and helical) structure with the propagation along the b axis below a characteristic temperature. At 3.8 GPa, a helical magnetic order appears below 208 K, which hosts the spins in the ac plane and the propagation along the b axis. The high-pressure magnetotransport measurements in FeSe up to ~ 15 GPa are reported. Above ~ 6 GPa the sudden enhancement of superconductivity ($T_c \leq 38.3$ K) accompanies a suppression of magnetic order. The obtained phase diagram highlights unique features of FeSe among iron-based superconductors. And effect of pressure on the several Eu compounds have been investigated as results of joint program.

13. Iron arsenides with three-dimensional FeAs layer networks: Ca$_{n(n+1)/2}$(Fe$_{1-x}$Pt$_x$)$_{(2+3n)}$P$_{tn(n+1)/2}$(n=2,3): N. Katayama, S. Onari, K. Matsubayashi, Y. Uwatoko and H. Sawa, Sci. Rep. 6 (2016) 39280(1-5).


**Noguchi group**

We have studied the membrane shape transformations by proteins. (1) tubulation and rupture by the absorption of banana-shaped proteins. (2) high-genus stomatocyte (nuclear envelope shape) constructed by osmotic pressure and pore-size constraint by nuclear pore complex. We have also proposed new force decomposition methods of multibody potentials to calculate local stress and found that the obtained local stress largely depends on the decompositions even when they satisfy the conservation of translational and angular momentum.


Materials Synthesis and Characterization group

1. \(^{1,2}\) Pressure-induced non-superconducting phase of \(\beta\text{-Na}_{0.33}\text{V}_{2}\text{O}_{5}\) and the mechanism of high-pressure phase transitions in \(\beta\text{-Na}_{0.33}\text{V}_{2}\text{O}_{5}\) and \(\beta\text{-Li}_{0.33}\text{V}_{2}\text{O}_{5}\) at room temperature: A. Grzechnik, Y. Ueda, T. Yamauchi, M. Hanfland, P. Herin, V. Potapkin and K. Friese, J. Phys.: Condens. Matter 28 (2016) 035401(1-9).


5. \(^{1,2}\) Weak ferromagnetic order breaking the threefold rotational symmetry of the underlying kagome lattice in \(\text{CdCu}_{3}(\text{OH})_{6}(\text{NO}_{3})_{2}\cdot\text{H}_{2}\text{O}\): R. Okuma, T. Yajima, D. Nishio-Hamane, T. Okubo and Z. Hiroi, Phys. Rev. B 95 (2017) 094427.


Neutron Science Laboratory

Shibayama group

Shibayama group has been exploring the structure and dynamics of soft matter, especially polymer gels, micelles, thermo-responsive polymers, and thermostets, utilizing a combination of small-angle neutron scattering (SANS), small-angle X-ray scattering (SAXS), and dynamic light scattering (DLS). The objectives are to elucidate the relationship between the structure and variety of novel properties/functions of polymer gels/resins. The highlights of 2016 include investigation of (1) solvated structure of thermo-responsive polymers in ionic liquid, (2) structure of amphiphilic conetworks, (3) crosslinking inhomogeneities of phenolic resins, (4) pressure response of thermoresponsive polymer in ionic liquid, and (5) dynamic light scattering microscopy of turbid systems, (6) probe-SAXS of hydrogels under elongation, and so on.


\(^{1}\) Joint research with outside partners.

Yoshizawa group
A systematic study on a family of Ce-based non-centrosymmetric heavy fermion compounds CeTSi3 (T=transition metal ions) were studied, and the M-T phase diagram was established for CePdSi3. It is found that CePdSi3 shows extremely complicated multi-metamagnetic transitions. In order to elucidate such complicated magnetic phase diagram and magnetic properties from a microscopic basis, the crystalline electric field levels were studied for T=Pd, Pt, and Rh compounds with use of inelastic neutron scattering measurements.


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, we have succeeded to measure synchrotron X-ray diffraction of glassy carbon disulfide (CS2), propane (CH3CH2CH3) and propene (CH3CHCH2). These glasses were prepared by vapor-deposition at 3 K by using a cryostat developed for in-situ X-ray diffraction experiments in SPring-8. It was found that the local structures of these simple molecular glasses are similar to those of their crystalline states. Another topic is that we found unusual glass transitions in liquid alkylated tetraphenylporphirins (3,5-C6C10TPP and 2,5-C6C10TPP); they appeared in a wide temperature range between 150 and 230 K. It is amazing that these large molecules (molecular mass: 2538) exist in liquid states at room temperature. We named these liquids “super-high entropy liquids”. Other than above topics, we have obtained neutron diffraction data of nanoparticles of PdPtD and PdRuD alloy hydride systems. The analysis is going on.

* Joint research among groups within ISSP.


10. 熱測定と中性子散乱の相補利用による新規物質研究: 山室 修, 熱測定 (2017), accepted for publication.


12. ガラス転移温度: 山室 修, 「化学便覧基礎編改訂第 6 版」, 10.15, 日本化学会編, (丸善出版, 2017), accepted for publication.

**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; magnetic order in a buckled honeycomb lattice antiferromagnet, spin model of O2-based magnet in a nanoporous metal complex, dielectric and magnetic properties in relaxor magnet, magnetic excitations in a quantum breathing pyrochlore antiferromagnet, and continuous control of local magnetic moment by applied electric field in a multiferroic material.


† Joint research with outside partners.


**International MegaGauss Science Laboratory**

**Takeyama group**

1000 T class electro-magnetic flux compression megagauss generators were reconstructed and completed. A peak magnetic field of 450 T is obtained by the magagauss generator with less than 1 MJ energy injection. Magnetization measurement techniques are still in progress in the single-turn coil magagauss generator system, and the measurements using a co-axial type self-compensated pick-up-coil up to 130 T, and using magneto-optical techniques up to 200 T are currently achieved with high reliability, at very low temperature around 5 K. The methods have been applied to investigate spin structures of frustrated magnetic materials, multi-ferro materials, and quantum spin systems, etc.


**Kindo group**

The specific heat measurements under long pulsed magnetic field up to 43.5 T has been open for the joint-use research. Repetition of the maximum field generation is limited to three times in a day due to a cooling time for the magnet. We are making new high repetitive magnet to carry out the measurements at higher frequency.

* Joint research among groups within ISSP.


6. †Origin of positive out-of-plane magnetoconductivity in overdoped Bi$_{1.6}$Pb$_{0.4}$Sr$_2$CaCu$_{1.96}$Fe$_{0.04}$O$_{8+\delta}$: T. Watanabe, T. Usui, S. Adachi, Y. Teramoto, M. M. Dobroka, I. Kakeya, A. Kondo, K. Kindo and S. Kimura, Phys. Rev. B 94 (2016) 174517.


9. †Various disordered ground states and 1/3 magnetization-plateau-like behavior in the S=1/2 Ti$^3+$ kagome lattice antiferromagnets Rb$_2$NaTi$_3$F$_{12}$, Cs$_2$NaTi$_3$F$_{12}$, and Cs$_2$KTi$_3$F$_{12}$: M. Goto, H. Ueda, C. Michioka, A. Matsuo, K. Kindo and K. Yoshimura, Phys. Rev. B 94 (2016) 104432.


† Joint research with outside partners.


Tokunaga group

Multiferroic materials have been extensively studied partly because of their possible application to the memory devices with low power consumption. Our careful experiments revealed non-volatile magnetoelectric memory effects in a well-known multiferroic material, BiFeO3. We found application of positive (negative) electric fields to a sample causes decrease (increase) in the resistance. This bipolar RRAM effect observed at room temperature will be useful for memory devices.


* Joint research among groups within ISSP.
Y. Matsuda group
Spin state degree of freedom of cobalt oxides has been attracting much attention of scientists for a long time. The electronic and structural properties as well as the magnetic properties sometimes change dramatically. LaCoO$_3$ is one of the canonical cobalt oxides and has been studied for more than half century owing to its peculiar properties. We have investigated the magnetization process of LaCoO$_3$ in high magnetic fields of up to 133 T at several temperatures. In addition to the previously known magnetization jump at around 65 T at low temperatures, a novel phase has been observed to appear in higher fields when temperature is higher than about 30 K. The phase boundary in field-temperature (B-T) plane indicates this phase has lower entropy than the low field phase. The transition field increases with temperature and becomes higher than 130 T at 105 K. The spatial ordered state of the different spin states, namely high spin (S=2), intermediate spin (S=1) and low spin (S=0) states, is suggested. From theoretical point of view, field-induced transition to an excitonic insulating state possibly explains the observed phenomenon. Another cobalt oxide (Pr$_{1-x}$Y$_x$)$_{0.7}$Ca$_{0.3}$CoO$_3$ has also been studied in ultrahigh fields of up to 140 T and found that the B-T phase diagram determined by the magnetization and electric resistivity is qualitatively different from that of LaCoO$_3$. We also studied the magnetic field induced phase transition of solid oxygen using irreversible heating effect. In SPring-8, using a pulsed magnet being combined with intense synchrotron x-rays, the valence state of Ce in the heavy fermion compound CeIrIn$_5$ was directly examined in high magnetic fields of up to 40 T at 2 K.


Center of Computational Materials Science
Akai group
(1) A scheme that combines the nonequilibrium Green’s function method with the Korringa–Kohn–Rostoker Green’s function method is proposed. The method is applied to Schottky junctions composed of an Al/GaN/Al trilayer. The results show that a Schottky barrier is formed at an undoped GaN and Al interface. The transport property of this system under various finite bias voltages is calculated. It is shown that the asymmetric behavior of electron transport against the direction of bias voltage occurs in this system, confirming the feature of rectification. (2) A classical spin model derived ab initio for rare-earth-based permanent magnet compounds is presented. Our target compound, NdFe$_{12}$N, is a material that goes beyond today’s champion magnet compound Nd$_2$Fe$_{14}$B in its intrinsic magnetic properties with a simpler crystal structure. Calculated temperature dependence of the magnetization and the anisotropy field agrees with the latest experimental results in the leading order. Having put the realistic observables under our numerical control, we propose that engineering 5d-electron-mediated indirect exchange coupling between 4f-electrons in Nd and 3d-electrons from Fe would most critically help enhance the material’s utility over the operation-temperature range. (3) As a collaboration with experimental group, we have developed the method to calculate resonant MOKEZ spectra and apply it to the recent experiments performed at SPring8 by Kubota, et al. The ab-initio calculations explain the experimental results successfully, opening up the possibility to further extend the method so as to cover other systems such as compounds, disordered alloys and also to apply the second-harmonic generation.


$^\dagger$ Joint research with outside partners.


Ozaki group

The X-ray photoelectron spectroscopy (XPS) has become one of the most important and widely used techniques in studying chemical composition and electronic states in the vicinity of surfaces of materials. In spite of the long history of XPS and its importance in materials science, a general method had not been developed so far to calculate the absolute binding energies for both insulators and metals, including multiple splittings due to chemical shift, spin-orbit coupling, and exchange interaction, on equal footing. In this study, we have developed a novel computational method to calculate absolute binding energies of core levels in metals and insulators, based on a penalty functional and an exact Coulomb cutoff method in a framework of the density functional theory. The spurious interaction of core holes between supercells is avoided by the exact Coulomb cutoff method, while the variational penalty functional enables us to treat multiple splittings due to chemical shift, spin-orbit coupling, and exchange interaction on equal footing, both of which are not accessible by previous methods. It is demonstrated that the absolute binding energies of core levels for both metals and insulators are calculated by the proposed method in a mean absolute (relative) error of 0.4 eV (0.16 %) for eight cases compared to experimental values measured with X-ray photoemission spectroscopy within a generalized gradient approximation to the exchange-correlation functional. We have applied the developed method to silicene grown on ZrB₂ substrate, which is one of fascinating two-dimensional materials discovered recently. It is found that the obtained binding energies of Si-2p states of the planar structure are in good agreement with the XPS data measured at the photon factory, KEK, resulting in a success of detailed analysis of buckling form of silicene.


* Joint research among groups within ISSP.
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.


11. \textsuperscript{†} 角度分解光電子分光による精密測定で解き明かす銅酸化物高温超伝導体の擬ギャップと超伝導ギャップの競合関係：近藤 猛, 竹内 恒博, 細木 純, 固体物理 \textbf{51} (2016) 203-221.


\textsuperscript{†} Joint research with outside partners.


I. Matsuda group

In 2016, we made large progress in research of the novel materials, especially "borophene". We directly observed its metallicity and, moreover, we discovered the Dirac Fermions. It is of note that the borophene we synthesized is the first metal allotrope of boron in a history of the condensed matter physics. Concerning our developments and experiments of the advanced spectrosopies, we successfully achieved in generating new techniques. One of them is to determine the element-specific complex permittivity using a fast-switching undulator that we have developed at SPring-8 BL07LSU. The other is to measure soft X-ray photoemission spectrum by introducing a high-frequency laser. The time-resolved data for joint-researches among groups within ISSP.


* Joint research among groups within ISSP.


† Joint research with outside partners.
Kobayashi group

We are developing a high-average-power, femtosecond laser system with Yb-doped fibers. Average power of 100 W is achieved.


11. ファイバーレーザー技術領域狭帯域 193nm 固体レーザーの開発：伊藤 紳二, 玄 洪文, 五十嵐 裕紀, 趙 智剛, 小林 洋平, 光学 第 46 巻第 4 号 (2017) 125-130.

Itatani group

First, we continued the development of intense ultrafast MIR sources based on optical parametric amplification (OPA). One source was BIBO-based OPA followed by difference frequency generation, which covered the spectral range in 5-10 μm. Another source was KTA-based OPA that produced 100-%μJ, 100-fs pulses at 3.2 μm. Both sources had stable carrier-envelope phases (CEPs), and were used to study high harmonic generation (HHG) in crystalline solids. Polarization-resolved measurement using an OPCPA-based infrared source, showing the capability of IR-OPCPA to extend the attosecond methodology from XUV (<200 eV) to soft-X-ray (>200 eV) regions. Such energy upscale is needed to realize ultrafast soft-X-ray spectroscopy in future. We also started to develop a novel velocity-map-imaging photoelectron spectrometer under the collaboration with Prof. Kling group in Germany. This apparatus is designed to measure the momentum distribution of photoelectrons up to 2 keV, which will be used to explore high-energy attosecond processes induced by long-wavelength optical fields.


* Joint research among groups within ISSP.


**Harada group**

Integrating the ambient pressure setup using a differential pumping system realized by FY2015, micro-focusing of the X-ray beam using KB mirrors, and angle resolved mode using the newly developed rotation chamber, we realized the following soft X-ray resonant inelastic X-ray scattering (RIXS) experiments in FY2016. (A)Observation of two-dimensional (excitation energy and momentum) mapping of RIXS By optimizing the focusing optics and using the horizontal rotation of the RIXS spectrometer, angle resolved soft X-ray RIXS spectra of high Tc cuprates were obtained across O 1s resonance, which provides unique information about the Cu-O mixing and resultant dispersion from the oxygen side. (B)Combined use of soft X-ray diffraction and RIXS using the angle resolved system High resolution angle resolved RIXS enables us combined use of soft X-ray diffraction from the elastic part and soft X-ray RIXS from the inelastic part. The first demonstration of such experiment was performed for LaSrFeO$_4$ which is well known to exhibit orbital ordering. Spatial modulation of the electron density yields not only soft X-ray diffraction but also intensity modulation of RIXS by the scattering angle, showing possibility of measuring ‘inelastic X-ray diffraction’. This year we have accepted 14 collaborative works at BL07LSU HORNET endstation, which include the above two topics and study on water encapsulated in polymers, bio-inspired materials, humidity dependence of functional transition metal complexes and operando analysis of Li ion battery electrodes.


† Joint research with outside partners.
Wadati group

We succeeded in the observation of photoinduced demagnetization and insulator-to-metal transition in ferromagnetic insulating BaFeO$_3$ thin films by time-resolved x-ray magnetic circular dichroism. We also studied the thickness-dependent physical properties of La$_{1/3}$Sr$_{2/3}$FeO$_3$ thin films to obtain critical thickness for charge ordering.


Kondo group

We use angle-resolved photoemission spectroscopy (ARPES) with ultrahigh energy resolution. The main findings in 2016 are as follows: (1) Slater to Mott Crossover in the Metal to Insulator Transition of Nd$_2$Ir$_2$O$_7$, (2) Orbital-Dependent Band Narrowing on the Topmost Layer of Sr$_2$RuO$_4$, and (3) Coherent control over three-dimensional spin polarization for the spin-orbit coupled surface state of Bi$_2$Se$_3$.


* Joint research among groups within ISSP.


**Okazaki group**

We have investigated superconducting-gap structures of unconventional superconductors such as iron-based superconductors and Bi\textsubscript{2}S\textsubscript{2}-based superconductors by a low-temperature and high-resolution laser ARPES apparatus and transient electronic structures in photo-excited non-equilibrium states by a time-resolved ARPES apparatus using EUV and SX lasers. In the fiscal year 2016, we have revealed unconventional superconductivity in Nd\textsubscript{0.71}F\textsubscript{0.29}Bi\textsubscript{2}S\textsubscript{2} from its quite anisotropic superconducting structures in photo-excited non-equilibrium states by a time-resolved ARPES apparatus using EUV and SX lasers. In the fiscal year 2016, we have revealed unconventional superconductivity in Nd\textsubscript{0.71}F\textsubscript{0.29}Bi\textsubscript{2}S\textsubscript{2} from its quite anisotropic superconducting gap structure, and importance of the spin-orbit coupling for the superconducting gap anisotropy of Ba(Fe\textsubscript{0.65}Ru\textsubscript{0.35})\textsubscript{2}As\textsubscript{2}.


5. Unconventional superconductivity in the Bi\textsubscript{2}S\textsubscript{2}-based layered superconductor Nd\textsubscript{0.71}F\textsubscript{0.29}Bi\textsubscript{2}S\textsubscript{2}: Y. Ota, K. Okazaki, H. Q. Yamamoto, T. Yamamoto, S. Watanabe, C. Chen, M. Nagao, S. Watauchi, I. Tanaka, Y. Takano and S. Shin, Phys. Rev. Lett 118 (2017) 167002(1-6).

† Joint research with outside partners.