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 SrCu2(BO3)2. (2) Microscopic examination by angle resolved 27Al-NMR of the quadrupole order and multipolar fluctuations in PrTi2Al20. (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.


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 UBe13 (Tc=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 Sr2RuO4 have been measured down to 100 mK under a precise control of the magnetic-field 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 Γ = ξ / μB of as large as 60 even at 100 mK. The present result raises fundamental issues in both the existing spin-triplet and spin-singlet scenarios.


* Joint research among groups within ISSP.


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$_2$(Cat-ETF-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)$_2$BF$_4$, and (3) to observe the anisotropic transport behavior under uniaxial strain in the pressure induced superconductor

† Joint research with outside partners.
β-(meso-DMBEDT-TTF)₂PF₆. The introduction of a large variety of molecule degree of freedom to solid promises the development of new trends in functional molecular materials.


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$_2$Al$_2$O$_9$ 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 PrV$_2$Al$_2$O$_9$ 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.


* Joint research among groups within ISSP.


17. Conduction electron spin resonance in the $\alpha$-Yb$_{1-x}$Fe$_x$AlB$_4$ ($0 \leq x \leq 0.50$) and $\alpha$-Lu$_x$AlB$_4$ 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.


† Joint research with outside partners.


**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, even if the orbital degeneracy is not taken into account.


**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 $r_p$ 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 $r_p \sim 20$, 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 $m^* \sim 0$, leading to a new filed of "light-fermion physics".


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* Joint research among groups within ISSP.


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.


Tsunetsugu group

We have studied an unidentified phase transition observed in the heavy fermion compound PrIr₂Zn₂₀. 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.


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.


* 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.

3. Joint research with outside partners.

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 magnetoresistance due to localization effect and oscillatory behavior attributed to geometrical resonance and observed.


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 hydrogenation has been detected through the inverse spin Hall effect.


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.


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.


3. Robust Protection from Backscattering in the Topological Insulator Bi1.5Sb0.5Te1.7Se1.3; S. Kim, S. Yoshizawa, Y. Iishida, 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.


Yoshinobu group

We conducted several research projects in the fiscal year 2014. (1) The activation and hydrogenation of \(\text{CO}_2\) 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 oxynitrile of SrTiO\(_3\).


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
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)-(√7 × √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.


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$_2$NiMnO$_6$ 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.


* Joint research among groups within ISSP.

Division of Physics in Extreme Conditions

Uwatoko group

The electrical resistivity of the nonmagnetic quadrupolar system PrTi$_2$Al$_{20}$ 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$_2$Te$_3$ 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 $\approx$ 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$\approx$2 K emerges at the critical pressure Pc$\approx$8 GPa, where the first-order antiferromagnetic transition at TN≈265 K under ambient pressure is completely suppressed.


† Joint research with outside partners.


**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 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.


**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$_2$Mn$_3$F$_{11}$. A kagome-type array of regular triangles composed of Mn$^2+$ ions (spin $5/2$) deforms so as to generate the next-nearest-neighbor interaction $J_2$ between three out of six spins in the hexagon of a normal kagome lattice, in addition to the nearest-neighbor interaction $J_1$. As a function of $J_2/J_1$, this lattice can interconnect the kagome ($J_2 = 0$) and the triangular ($J_2 = J_1$) lattices and thus is called the kagome–triangular (KT) lattice. Magnetic susceptibility and heat capacity measurements performed on a polycrystalline sample of NaBa$_2$Mn$_3$F$_{11}$ 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 $J_1$ and antiferromagnetic $J_2$. NaBa$_2$Mn$_3$F$_{11}$ may actually exist in this regime according to the results of our analysis based on classical Monte-Carlo simulation. In a frustrated $J_1$–$J_2$ chain with the nearest-neighbor ferromagnetic interaction $J_1$ and the next-nearest-neighbor antiferromagnetic interaction $J_2$, 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 CuO$_2$ chains, is a good candidate $J_1$–$J_2$ chain antiferromagnet. The exchange interactions are estimated as $J_1 = -51$ K and $J_2 = 36$ 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.

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


**Kawashima group**

We have been investigating quantum spin/boson systems and frustrated systems by means of large-scale numerical simulation. 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.


* 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 biomembranes and dynamics of deformable microsimmers.

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.

Materials Synthesis and Characterization group


† Joint research with outside partners.

9. Electrochemical properties of LiMn$_2$Fe$_{1-x}$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.


**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.


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-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.

4. Neutron Diffraction Study of Parasitic Nd-Moment Order in the Checkerboard-Type Phase Nd$_{1.5}$Sr$_{0.7}$NiO$_4$: R.

† Joint research with outside partners.

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-PdH 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.


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 Rb2Cu2Mo3O12, spin gap in antiferromagnetic alternating spin 3/2 chain substances RCrGeO5 (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.


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. We have developed a calculation code, which is used for dynamical simulation of the magnetic-flux compression. This phase is very similar to that discovered in other chromium spinel oxide, CdCr$_2$O$_4$, and ZnCr$_2$O$_4$. We have found that relative ordering between optical bright and dark band-edge excitons is chirality dependent in semiconductors single wall carbon nanotubes, and a bright exciton can stay at the lowest energy for a certain chirality carbon nanotube.


† Joint research with outside partners.

**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.

13. † Unconventional spin freezing in the highly two-dimensional spin-1/2 kagome antiferromagnet

* Joint research among groups within ISSP.


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.


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 YbAlB₄ and CeRh₂Si₂ 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$_2$Si$_2$, 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.


**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$_3$Fe$_{17}$B and Sm$_2$Fe$_{17}$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-electron 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-equilibrium 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.


* Joint research among groups within ISSP.


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.


† 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.


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.


* Joint research among groups within ISSP.


8. †Robust Protection from Backscattering in the Topological Insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.5}\text{Se}_{1.5}$: 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).


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$_2$Se$_3$ thin-films were investigated by surface X-ray diffraction and magnetotransport measurements. It is reported that Cu-doped Bi$_2$Se$_3$ 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.


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.


* Joint research among groups within ISSP.
using VUV and SX rays. Using a seeded free-electron laser at FERMI@ELETTRA in Italy, we have succeeded in tracing the development of the rotating analyzer ellipsometry method for measuring the resonant magneto-optical Ker effect (RMOKE) supported picosecond-time-resolved SX photoemission spectroscopy experiments of joint researches. Studies of photovoltaics type undulator in the electron storage ring to realize fast switching of the light polarizations. At the end-station, we routinely soft X-rays (SX). At SPring-8 BL07LSU, we have adjusted phase shifters with electromagnetic coils of the segmented cross-I. Matsuda group

† Joint research with outside partners.

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.


**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.


2. **Wavefront analysis of high-efficiency, large-scale, thin transmission gratings:** C. ZHOU, T. SEKI, T. KITAMURA, Y.

* Joint research among groups within ISSP.


**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 VO2 and graphene were successfully performed.


9. Ultrabroadband infrared chirped mirrors characterized by a white-light Michelson interferometer: K. Kaneshima, M.
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$_2$O ice under the pressure around $10^5$ Pa. We have also 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$_3$; v) Distinguishing between High- and Low-Spin States for Divalent Mn in Mn-Based Prussian Blue Analogue by High-Resolution Soft X-ray Emission Spectroscopy; vi) 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; vii) Development of 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. Kadono, T. Okane, Y. Saitoh, H. Yamagami, Y. Harada, M. Oshima, M. Tanaka and A. Fujimori, Appl. Phys. Lett. 105 (2014) 032403(1-4).


Wadati group

Our main experimental techniques are synchrotron-based x-ray spectroscopy and scattering. We studied the orbital and magnetic phase transitions in Pr₀.₅Ca₀.₅MnO₃/La₀.₅Ca₀.₅MnO₃ 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 Pr₀.₅Ca₀.₅MnO₃ thin films by using x-ray free electron laser in LCLS (USA), and observed a time-dependent order parameter by creating coherent phonons.


† Joint research with outside partners.

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 $T_c$ in the pnictide high-$T_c$ superconductor.


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.


* Joint research among groups within ISSP.