ISSP 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 2011 include: (1) Investigation in high magnetic field of anisotropic spin dynamics of $LiCuVO_4$, a quasi 1D frustrated antiferromagnet with a ferromagnetic nearest neighbor interaction, which enabled us to find evidence for the two-magnon bound state in the field range where the spin density wave order takes place at low temperatures, (2) Srearch for a spin nematic order in $LiCuVO_4$ in very high magnetic fields immediately below the saturation, (3) Detailed and systematic NMR experiments on the distorted Kagome spin system volborthite, which allowed us to determine the precise phase diagram with unusual spin order and fluctuation effects in high magnetic fields.

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6. 容積効率にこだわった高圧セルによる 10GPa 級 NMR 測定: 北川健 太郎, 松林 和幸, 後藤 弘匡, 松本 武彦, 上床 美也, 八木 健彦, 瀧川 仁, 高圧力の科学と技術 **22** (2012) 198-205.

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 2012. (1) Field and temperature variations of the specific heat $C(H,T)$ of the heavy fermion superconductor $CeCu_2Si_2$ (T_c=0.6 K) were examined at temperatures down to 50 mK. Quite unexpectedly, the low temperature $C(H,T)$ indicates an exponential decay with a two-gap feature in its temperature variation, along with a linear dependence as a function of H. The results strongly indicate that the superconducting gap is fully opened, in sharp contrast to the general belief that $CeCu_2Si_2$ is a d-wave superconductor. (2) We examined the low temperature magnetization of the Yb dimer compound Yb_2Pt_3Pb. The results confirm the model, proposed in a preceding study by Ochiai et al., in which Yb^{3+} has an Ising-type Kramers doublet in the crystalline electric field ground state. Interestingly, the system exhibits a spin-flop like phase at low temperatures and in high fields. This unusual behavior of the magnetization is explained by assuming a pseudo-spin flop of the doublet, which can be stabilized by introducing a high-rank multipole interaction. (3) We measured the magnetization of the S=1/2 one dimensional Heisenberg antiferromagnet CuPzN (interaction parameter $J\sim$10 K) at temperatures down to 80 mK in magnetic fields up to 15 T. At the base temperature (T/J=0.008), the field variation of the magnetization is found to closely follow the exact solution obtained by the Bethe ansatz.

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Mori group

We have successfully developed and characterized the functional molecular materials. The major achievements in 2012 are (1) to develop the purely organic single-component organic conductor kappa-H_3(Cat-EDT-ST)$_2$ which shows metallic state under 1GPa, (2) to develop the proton-electron coupled conductor with the proton arrangement driven by the charge ordering, and (3)to develop the pressure-induced superconductor beta-(meso-DMBEDT-TTF)$_2AsF_6$ which shows the checkerboard-type charge ordering at ambient pressure.

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Tajima group

Our main subject is the electrical properties on molecular assemblies especially on organic thin films and conducting molecular crystals. The major achievements in 2012 include (1) spin injection experiments into a organic conductive polymer PEDOT:PSS, (2) a detailed report on a new technique for determining a trap density function based on the photo-CELIV measurements, and (3) magnetic torque studies on conducting phthalocyanine salts.

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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 2012. (1) We have found that Pr$_2$Zr$_2$O$_7$, the sister insulating compound of Pr$_2$Ir$_2$O$_7$, has strong quantum monopolar fluctuations in the spin ice state. This suggests the monopole in this exchange-based spin ice may propagate coherently in the lattice, in contrast with the diffusive motion of the classical counterpart. (2) We discovered heavy fermion superconductivity in the ferroquadrupolar state in the quadrupolar Kondo lattice system PrTi$_2$Al$_{20}$. (3) The strong temperature and field dependence of the Hall resistivity of β-YbAlB$_4$ indicates that the low coherence temperature of 40 K in comparison with the valence fluctuation scale of 300 K. The results further suggest the band dependence of the localized and itinerant characters of *f*-electrons.

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28. Quantum fluctuations in spin-ice-like $Pr_2Zr_2O_7$: K. Kimura, S. Nakatsuji, J. J. Wen, C. Broholm, M. B. Stone and E. Nishibori & H. Sawa, Nature Commun. **4** (2013) 1-6, in print.

29. 銅酸化物における乱れに強い量子液体状態: 中辻 知, 澤 博, 「超伝導現象と高温超伝導体」, 新日本編集企画, (NTS 出版社, 2013), 475-481.

Ohgushi group

Our group is focused on an exploratory synthesis and characterization of oxides, chalcogenides, and intermetallics. The major achievements in the fiscal year 2012 are (1) investigation of electronic properties of Fe-based ladder compounds, and (2) elucidation of magnetic and orbital structures of 5*d* transition metal oxides by means of resonant x-ray diffraction.

1. †*Abrupt change in the energy gap of superconducting $Ba_{1-x}K_xFe_2As_2$ single crystals with hole doping: W. Malaeb, T. Shimojima, Y. Ishida, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, T. Saito, C. H. Lee, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, Y. Kohori, A. Iyo, H. Eisaki, C. -T. Chen, S. Watanabe, H. Ikeda and S. Shin, Phys. Rev. B **86** (2012) 165117 (1-7).

2. *Block magnetism coupled with local distortion in the iron-based spin-ladder compound $BaFe_2Se_3$: Y. Nambu, K. Ohgushi, S. Suzuki, F. Du, M. Avdeev, Y. Uwatoko, K. Munakata, H. Fukazawa, S. Chi, Y. Ueda and T. Sato, Phys. Rev. B **85** (2012) 064413(1-5).

3. Doping dependence of Hall coefficient and evolution of coherent electronic state in the normal state of the Fe-based superconductor $Ba_{1-x}K_xFe_2As_2$: K. Ohgushi and Y. Kiuchi, Phys. Rev. B **85** (2012) 064522(1-5).

4. *Stripelike magnetism in a mixed-valence insulating state of the Fe-based ladder compound $CsFe_2Se_3$: F. Du, K. Ohgushi, Y. Nambu, T. Kawakami, M. Avdeev, Y. Hirata, Y. Watanabe, T.-J. Sato and Y. Ueda, Phys. Rev. B **85** (2012) 214436(1-5).

5. *Tetrahedral Magnetic Order and the Metal-Insulator Transition in the Pyrochlore Lattice of $Cd_2Os_2O_7$: J. Yamaura, K. Ohgushi, H. Ohsumi, T. Hasegawa, I. Yamauchi, K. Sugimoto, S. Takeshita, A. Tokuda, M. Takata, M. Udagawa, M. Takigawa, H. Harima, T. Arima and Z. Hiroi, Phys. Rev. Lett. **108** (2012) 247205(1-5).

6. *Insitu observation of shear stress-induced perovskite to post-perovskite phase transition in $CaIrO_3$ and the development of its deformation texture in a diamond-anvil cell up to 30 GPa: K. Niwa, N. Miyajima, Y. Seto, K. Ohgushi, H. Gotou and T. Yagi, Phys. Earth Planet. Inter. **194–195** (2012) 10–17.

7. *Observation of Phonon-Assisted Magnon Absorption in Spin–Orbit Coupling Induced Mott Insulator Sr_2IrO_4: Y. Hirata, H. Tajima and K. Ohgushi, J. Phys. Soc. Jpn. **82** (2013) 035002(1-2).

8. Complex orbital state stabilized by strong spin-orbit coupling in a metallic iridium oxide IrO_2: Y. Hirata, K. Ohgushi, J.-I. Yamaura, H. Ohsumi, S. Takeshita, M. Takata and T. Arima, Phys. Rev. B **87** (2013) 161111(1-5).

9. Magnetoelasticity in ACr_2O_4 spinel oxides (A = Mn, Fe, Co, Ni, and Cu): V. Kocsis, S. Bordács, D. Varjas, K. Penc, A. Abouelsayed, C. A. Kuntscher, K. Ohgushi, Y. Tokura and I. Kézsmárki, Phys. Rev. B **87** (2013) 064416(1-9).

10. *Mechanism of Enhanced Optical Second-Harmonic Generation in the Conducting Pyrochlore-Type $Pb_2Ir_2O_{7-x}$ Oxide Compound: Y. Hirata, M. Nakajima, Y. Nomura, H. Tajima, Y. Matsushita, K. Asoh, Y. Kiuchi, A. G. Eguiluz, R. Arita, T. Suemoto and K. Ohgushi, Phys. Rev. Lett. **110** (2013) 187402(1-5).

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13. †*B*-Site Deficiencies in *A*-site-Ordered Perovskite $LaCu_3Pt_{3.75}O_{12}$: M. Ochi, I. Yamada, K. Ohgushi, Y. Kusano, M. Mizumaki, R. Takahashi, S. Yagi, N. Nishiyama, T. Inoue and T. Irifune, Inorg. Chem. **52** (2013) 3985-3989.

14. †Pd^{2+}-Incorporated Perovskite $CaPd_3B_4O_{12}$(*B* = Ti, V): K. Shiro, I. Yamada, N. Ikeda, K. Ohgushi, M. Mizumaki, R. Takahashi, N. Nishiyama, T. Inoue and T. Irifune, Inorg. Chem. **52** (2013) 1604-1609.

Division of Condensed Matter Theory

K. Ueda group

Following the previous year, the problem of the Kondo effect of a magnetic ion vibrating in a metal was investigated. In 2011, we investigated the nature of Kondo effect in the weak electron correlation regime from a different point of view. It has turned out that the essential physics in this regime can be understood by the concept of electric dipolar Kondo effect. As another topic, our group focused on shot noise observed in the Kondo tunnelingthrough the dot with orbital degeneracy. The Fano factor determined by the Wilson ratio was calculated by employing the NRG calculation. This approach was also applied to the FQH edge states. Analyzing shot noise at finite temperatures, we proposed a way to observe the fractional statistics for Laughlin quasi-particles.

1. Dimensional Reduction and Odd-Frequency Pairing of the Checkerboard-Lattice Hubbard Model at 1/4-Filling: Y. Yanagi, Y. Yamashita and K. Ueda, J. Phys. Soc. Jpn. **81** (2012) 123701(1-4).

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3. 固体物理特集号「重い電子系の物理の最近の発展」 はじめに: 上田 和夫, 固体物理 **67** (2012) 509-510.

4. 高温超伝導–理論–: 上田 和夫, 「物性物理学ハンドブック」, 川畑有郷・鹿児島誠一・北岡良雄・上田正仁, (朝倉書店, 2012), 175-182.

Takada group

Employing several techniques including the Green's-function approach, the density-matrix renormalization group, quantum Monte Carlo simulations, band-structure calculations, 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 studied the following issues: (1) With use of both the diffusion Monte Carlo method and the density functional theory, a detailed analysis is made on the system of a proton embedded in an electron gas with a view of clarifying the difference between this first-principles system and the impurity Anderson model devised for investigating the Kondo physics. With the decrease of the electron density of the electron gas, the electronic state around the proton changes in three steps, i.e., from the hydron H^+ with itinerant metallic screening to the totally localized hydride H^- through the Kondo-resonant nonmagnetic neutral hydrogen H. The transition of each step is abrupt. (2) With use of analytical methods as well as the powerful self-consistent numerical scheme of GWΓ, we have discovered an electron-like elementally excitation (pseudoelectron) in the Luttinger liquid. The pseudoelectron is considered as a spinon-(anti)holon composite particle. Although it manifests itself only as the small cusp behavior in the one-electron spectral function near the Fermi level, it constitutes the main structure if the momentum is far away from the Fermi momentum. (3) With a better functional form for Γ always satisfying not only the Ward identity but also the momentum conservation law, we have investigated the low-density electron gas in the GWΓ scheme to find an anomalous mass reduction as a result of avoiding the collapse of the normal state into a spontaneously excited electron-hole liquid (excitonic liquid).

1. *Coulomb Frustrated Phase Separation in Quasi-Two-Dimensional Organic Conductors on the Verge of Charge Ordering: K. Yoshimi and H. Maebashi, J. Phys. Soc. Jpn. **81** (2012) 063003(1-4).

2. Superconductivity in a Correlated E ⊗ e Jahn-Teller System: C. Hori, H. Maebashi and Y. Takada, J. Supercond. Nov. Magn. **25** (2012) 1369-1373.

3. 超伝導転移温度の第一原理計算: 高田 康民, 「岩波講座：計算科学　「計算と物質」」, 第８章, 押山淳, (岩波書店, 2012), 221-267.

Oshikawa group

We studied a wide range of fundamental problems in condensed matter theory and statistical mechanics. In particular, we discussed doping of holes into the spin liquid phases of the Quantum Dimer Model (QDM). A fundamental issue is the possible existence of a superconducting phase in such systems and its properties. For this purpose, the question of the statistics of the mobile holes (or "holons") was addressed first. We proved a general "statistical transmutation" symmetry of such doped QDM by using composite operators of dimers and holes. This exact transformation enables to define duality equivalence classes (or families) of doped QDM, and provides the analytic framework to analyze dynamical statistical transmutations. We then discussed possible superconducting phases of the system. In particular, the possibility of an exotic superconducting phase originating from the condensation of (bosonic) charge-e holons was examined. It was shown that flux quantization does not distinguish such an exotic superconducting phase from a standard one due to Cooper pairing. Thus we proposed a new gauge-invariant holon Green's function, as a mean to detect the exotic phase. A numerical evidence supported its existence in the doped QDM on a triangular lattice.

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2. †Field theory analysis of S=1 antiferromagnetic bond-alternating chains in the dimer phase: J. Tamaki and M. Oshikawa, Phys. Rev. B **85** (2012) 134431(1-10).
3. †General method for calculating the universal conductance of strongly correlated junctions of multiple quantum wires: A. Rahmani, C.-Y. Hou, A. Feiguin, M. Oshikawa, C. Chamon and I. Affleck, Phys. Rev. B **85** (2012) 045120(1-24).
4. Mass ratio of elementary excitations in frustrated antiferromagnetic chains with dimerization: S. Takayoshi and M. Oshikawa, Phys. Rev. B **86** (2012) 144408 (1-6).
5. †Quasiparticle statistics and braiding from ground-state entanglement: Y. Zhang, T. Grover, A. Turner, M. Oshikawa and A. Vishwanath, Phys. Rev. B **85** (2012) 235151(1-15).
6. †Symmetry protection of topological phases in one-dimensional quantum spin systems: F. Pollmann, E. Berg, A. M. Turner and M. Oshikawa, Phys. Rev. B **85** (2012) 075125(1-9).
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8. †Boundary Resonances in S=1/2 Antiferromagnetic Chains Under a Staggered Field: S. C. Furuya and M. Oshikawa, Phys. Rev. Lett. **109** (2012) 247603(1-5).
9. †Electron Spin Resonance Shift in Spin Ladder Compounds: S. C. Furuya, P. Bouillot, C. Kollath, M. Oshikawa and T. Giamarchi, Phys. Rev. Lett. **108** (2012) 037204(1-5).
10. †Instability in Magnetic Materials with a Dynamical Axion Field: H. Ooguri and M. Oshikawa, Phys. Rev. Lett. **108** (2012) 161803(1-5).
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12. †Electron spin resonance shifts in S=1 antiferromagnetic chains: S. C. Furuya, Y. Maeda and M. Oshikawa, Phys. Rev. B **87** (2013) 125122 (1-10).
13. †Hole statistics and superfluid phases in quantum dimer models: C. A. Lamas, A. Ralko, M. Oshikawa, D. Poilblanc and P. Pujol, Phys. Rev. B **87** (2013) 104512(1-20).

Tsunetsugu group

We have investigated p-wave superconductivity near a transverse saturation field in easy-axis ferromagnets and demonstrated that soft-magnon excitations lead to enhancement and re-entrance of the superconducting transition temperature. This explains the enhanced superconducting transition temperature in the reentrant superconductivity in URhGe under transverse magnetic fields.

We have also extended our earlier analysis about phonon coupled Kondo systems. We have constructed a boundary conformal field theory (BCFT) with non-magnetic SO(5) degrees of freedom, which successfully explains earlier numerical results. We have further developed a powerful continuous-time quantum Monte Carlo algorithm for general phonon-assisted hybridization Anderson models and checked the validity of the results by the BCFT.

We have also clarified various details of transport criticality in electric conductivity near the Mott transition in a frustrated Hubbard model, using large-scale numerical simulations based on the cluster dynamical mean-field theory.

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2. Transport Criticality in Triangular Lattice Hubbard Model: T. Sato, K. Hattori and H. Tsunetsugu, J. Phys. Soc. Jpn. **81** (2012) 083703 (4 pages).

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4. Conventional and charge-six superfluids from melting hexagonal Fulde-Ferrell-Larkin-Ovchinnikov phases in two dimensions: D. F. Agterberg, M. Geracie and H. Tsunetsugu, Phys. Rev. B **84** (2012) 014513 (7 pages).

5. Non-Fermi liquid, unscreened scalar chirality, and parafermions in a frustrated tetrahedron Anderson model: K. Hattori and H. Tsunetsugu, Phys. Rev. B **86** (2012) 054421 (6 pages).

6. Transport criticality at the Mott transition in a triangular-lattice Hubbard model: T. Sato, K. Hattori and H. Tsunetsugu, Phys. Rev. B **86** (2012) 235137 (13 pages).

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8. Exotic disordered phases in the quantum J1-J2 model on the honeycomb lattice: H. Zhang and C. A. Lamas, Phys. Rev. B **87** (2013) 024415 (10 pages).

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Kohmoto group

Twisted bilayer graphen is studied. Especially energy versus magnetic field shows a fractal Hofstadter butterfly. Since supercell is large one can expect nontrivial Hall conductance (TKNN integer) may be observed in a high magnetic field experiment. Also edge states at the interface between monolayer and bilayer are studied and its properties of applied electric field is clarified. Fibonacci optical lattice are studied and it is shown that soliton states and critical states can coexist multifractal analysis.

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2. Multifractals Competing with Solitons on Fibonacci Optical Lattices: M. Takahashi, H. Katsura, M. Kohmoto and T. Koma, New J. Phys. **14** (2012) 113012-113027.

3. Time-Reversal Symmetry in Non-Hermitian Systems: M. Sato, K. Hasebe, K. Esaki and M. Kohmoto, Prog. Theor. Phys. **127** (2012) 937-974.

4. Theory of tunneling conductance and surface-state transition in superconducting topological insulators: A. Yamakage, K. Yada, M. Sato and Y. Tanaka, Phys. Rev. B **85** (2012) 180509(R)(1-5).

5. Topological Field Theory for p-wave Superconductors: T. H. Hansson, A. Karlhede and M. Sato, New J. Phys. **14** (2012) 063017(10pages).

6. Symmetry and Topology in Superconductors —Odd-Frequency Pairing and Edge States—: Y. Tanaka, M. Sato and N. Nagaosa, J. Phys. Soc. Jpn. **81** (2012) 011013(1-34).

Sugino group

The activity of Sugino group consists of (1) investigation of the property of materials related to energy-conversion, (2) developing density functional methods for computing dynamical process in matters, and (3) developing a many-body scheme to calculate the ground-state and the excited-state energies. Particularly, there was breakthrough in the algorithm of handling the many-body wavefunctions, possibly leading to one of the post density functional theories (post-DFT).

1. Possible magnetic behavior in oxygen-deficient β-PtO_2: Y. Yang, O. Sugino and T. Ohno, Phys. Rev. B **85** (2012) 035204(1-13).

2. First-Principles Molecular Dynamics at a Constant Electrode Potential: N. Bonnet, T. Morishita, O. Sugino and M. Otani, Phys. Rev. Lett. **109** (2012) 266101(1-5).

3. Symmetric Tensor Decomposition Description of Fermionic Many-Body Wave Functions: W. Uemura and O. Sugino, Phys. Rev. Lett. **109** (2012) 253001(1-5).

4. A GW+Bethe-Salpeter calculation on photoabsorption spectra of $(CdSe)_3$ and $(CdSe)_6$ clusters: Y. Noguchi, O. Sugino, M. Nagaoka, S. Ishii and K. Ohno, J. Chem. Phys. **137** (2012) 024306 (1-5).

5. Analytical expression for the excited-state force from density-functional perturbation theory: T. Tsukagoshi and O. Sugino, Phys. Rev. A **86** (2012) 064501(1-4).

6. Quantum dissipative dynamics using the Doebner–Goldin equation: T. Tsukagoshi and O. Sugino, Physics Letters A **376** (2012) 3033-3037.

7. Band gap of β-PtO_2 from first-principles: Y. Yang, O. Sugino and T. Ohno, AIP Advances **2** (2012) 022172(1-7).

8. The charged interface between Pt and water: First principles molecular dynamics simulations: T. Ikeshoji, M. Otani, I. Hamada, O. Sugino, Y. Morikawa, Y. Okamoto, Y. Qian and I. Yagi, AIP Advances **2** (2012) 032182(1-10).

9. Nonadiabatic couplings from time-dependent density functional theory: Formulation by the Kohn-Sham derivative matrix within density functional perturbation theory: C. Hu, T. Tsukagoshi, O. Sugino and K. Watanabe, Phys. Rev. B **87** (2013) 035421(1-7).

10. Electronic structures of oxygen-deficient Ta_2O_5: Y. Yang, H.-H. Nahm, O. Sugino and T. Ohno, AIP Advances **3** (2013) 042101(1-8).

Kato group

The main research subject in our laboratory is theory of nonequilibrium properties in nanoscale devices. We have studied nonequilibrium noise of the Kondo regime in quantum dot systems by considering effect of Hund's exchange interaction, and purity of single photons in semiconductor microcavity. Dielectric properties in ferroelectric relaxors made from perovskite oxides have been studied by the Monte Carlo method. We have also studied spin diffusion length in normal metals in the context of anti-localization analysis in collaboration with Otani group.

1. *Coulomb Frustrated Phase Separation in Quasi-Two-Dimensional Organic Conductors on the Verge of Charge Ordering: K. Yoshimi and H. Maebashi, J. Phys. Soc. Jpn. **81** (2012) 063003(1-4).

2. Full Counting Statistics for Orbital-Degenerate Impurity Anderson Model with Hund's Rule Exchange Coupling: R. Sakano, Y. Nishikawa, A. Oguri, A. C. Hewson and S. Tarucha, Phys. Rev. Lett. **108** (2012) 266401(1-5).

3. Properties of a Single Photon Generated by a Solid-State Emitter: Effects of Pure Dephasing: E. Iyoda, T. Kato, T. Aoki, K. Edamatsu and K. Koshino, J. Phys. Soc. Jpn. **82** (2013) 014301(1-10).

4. Relaxor Behavior and Morphotropic Phase Boundary in a Simple Model: Y. Tomita and T. Kato, J. Phys. Soc. Jpn. **82** (2013) 063002(1-5).

5. 量子ドットの近藤効果による非平衡電流の完全係数統計: 阪野 塁, 小栗 章, 小林 研介, 固体物理 **47** (2013) 475-485.

Division of Nanoscale Science

Iye group

Thermoelectric effect in GaAs/AlGaAs 2DEG in the quantum Hall regime is studied by using a Corbino geometry. Thermopower generally contains contributions from two distinct mechanisms: diffusion and phonon drag. In order to selectively extract the diffusion contribution, microwave-heating technique is used to raise the electron temperature with the lattice temperature kept at the bath temperature. The measurement of the diagonal thermpower S_{rr} provides a unique opportunity to probe the entropy of the system in the QH plateau regions. The measured S_{rr} takes large values on the order of 1 meV/K in the regions where $\sigma_{rr} = 0$, alternating the sign at exact even integer fillings, in accordance with the theoretical prediction.

1. *Geometric resonances in the magnetoresistance of hexagonal lateral superlattices: Y. Kato, A. Endo, S. Katsumoto and Y. Iye, Phys. Rev. B **86** (2012) 235315(1-10).

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3. *Novel blockade due to spin-filtering with spin-orbit interaction: S. W. Kim, Y. Hashimto, Y. Iye and S. Katsumoto, J. Phys.: Conf. Ser. **400** (2012) 042032(1-4).

4. *Suppression of Andreev current due to transverse current flow in an InAs two-dimensional electrons: Y. Takahashi, Y. Hashimoto, Y. Iye and S. Katsumoto, Journal of Crystal Growth **378** (2013) S0022024813000705(1-4).

5. *Control of magnetic anisotropy in (Ga,Mn)As with etching depth of specimen boundaries: Y. Hashimoto, Y. Iye and S. Katsumoto, J. Cryst. Growth **378** (2013) S0022024812008329(1-4).

6. Commensurability oscillations in the rf conductivity of unidirectional lateral superlattices: measurement of anisotropic conductivity by coplanar waveguide: A. Endo, T. Kajioka and Y. Iye, J. Phys. Soc. Jpn **82** (2013) 054710(1-7).

7. Diffusion Thermopower of Quantum Hall States Measured in Corbino Geometry: S. Kobayakawa, A. Endo and Y. Iye, J. Phys. Soc. Jpn **82** (2013) 053702(1-4).

Katsumoto group

We have investigated the supercurrent through semiconductor 2-dimensional systems by applying spin current created with the spin Hall effect. We found the superconductivity established through the formation of the Andreev bound states is easily broken by small amount of spin current. This phenomenon seems to be promising in the application to low energy consumption logic devices.

1. Evidence of Spin-Filtering in Quantum Constrictions with Spin-Orbit Interaction: S. W. Kim, Y. Hashimoto, Y. Iye and S. Katsumoto, J. Phys. Soc. Jpn. **81** (2012) 054706 (1-5).

2. Detection of spin polarization utilizing singlet and triplet states in a single-lead quantum dot: T. Otsuka, Y. Sugihara, J. Yoneda, S. Katsumoto and S. Tarucha, Phys. Rev. B **86** (2012) 081308(1-4).

3. *Geometric resonances in the magnetoresistance of hexagonal lateral superlattices: Y. Kato, A. Endo, S. Katsumoto and Y. Iye, Phys. Rev. B **86** (2012) 235315(1-10).

4. *Magnetization dependent rectification in (Ga,Mn)As tri-layer tunnel junctions: Y. Hashimoto, H. Amano, Y. Iye and S. Katsumoto, J. Phys.: Conf. Ser. **400** (2012) 042016(1-4).

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6. Robustness of spin filtering against current leakage in a Rashba-Dresselhaus-Aharonov-Bohm interferometer: S. Matityahu, A. Aharony, O. Entin-Wohlman and S. Katsumoto, Phys. Rev. B **87** (2013) 205438(1-8).

7. *Suppression of Andreev current due to transverse current flow in an InAs two-dimensional electrons: Y. Takahashi, Y. Hashimoto, Y. Iye and S. Katsumoto, Journal of Crystal Growth **378** (2013) S0022024813000705(1-4).

8. *Control of magnetic anisotropy in (Ga,Mn)As with etching depth of specimen boundaries: Y. Hashimoto, Y. Iye and S. Katsumoto, J. Cryst. Growth **378** (2013) S0022024812008329(1-4).

9. Energy level spectroscopy of a quantum dot with a side‐coupled satellite dot: S. W. Kim, Y. Kuwabara, T. Otsuka, Y. Iye and S. Katsumoto, in: *AIP Conference Proceedings*, edited by Jisoon Ihm, Hyeonsik Cheong (American Institute of Physics, 2012), 393-394.

Otani group

This year we put our focus on the following three topics including the skew scattering mechanism of spin Hall effects, spin relaxation mechanism in lateral spin valve structures, and collective dynamics of coupled magnetic vortices. Concerning the first topic, we have performed collaborative research with German and French groups to find best combination of materials to induce large spin Hall effects originating from the skew scattering mechanisms. Based on these theoretical studies we have succeeded in demonstrating the giant spin Hall effect by doping copper with small amount of bismuth. Apart from the skew scattering studies, we have shown for the first time a possibility to use spin Hall effect as a means to detect higher order spin fluctuation. As to the second topic on the lateral spin valve, we have found the contribution of the surface spin flip scattering was non-negligible at low temperatures and was suppressed significantly by the surface capping layer of MgO. Thereby we were able to clarify that the spin relaxation process was well described in the framework of Elliot-Yafet mechanism. Furthermore we have demonstrated the non-local spin valve effect in the lateral spin valve structure consisting of silicon nano-wire and permalloy. As to the third topic on vortex dynamics, our final goal is to realize magnonic crystals. For this, we have been studying collective spin dynamics mediated by the dynamic dipolar interaction. When the nano-scale ferromagnetic

elements are very closely spaced with a separation of 50 nm, a gradual transition from completely uniform collective regime to a completely non-collective regime was observed as the azimuthal angle varies from 0° to 45°.

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Komori group

Electronic and atomic structures of the Au-adsorbed Ge(111) surface are studied by ARPES and STM. A triangle structure, which is mobile at room temperature on the surface, selectively dopes the electron-like metallic surface band. A band gap over 0.3 eV was found by ARPES in a regular array of the graphene nanoribbons that were formed selectively on the terrace of vicinal SiC(0001) surfaces by molecular beam epitaxy. The width of each nanoribbon is 10 nm. The gap size increases with decreasing the width of the nanoribbons. Regular arrays of square CrN islands were thermally fabricated on the Cu(001) surface. A short-range attractive interaction among the islands is the origin of the self-assembly.

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Yoshinobu group

We conducted several research projects in the fiscal year 2012. (1) The kinetic and geometric isotope effects and energy-level alignment of cyclohexane on clean and H-preadsorbed Rh(111) surfaces using IRAS, TPD, STM, SPA-LEED and SR-PES. (2) The adsorption states of CO_2 on Cu(997) studied by SR-PES and IRAS. (3) The thin file growth and electronic states of pentacene film on chemically modified Si(100) surfaces using PES and 4-probe surface conductivity measurements. (4) Spectroscopic characterization and transport properties of aromatic monolayers covalently attached to Si(111) surfaces by wet-chemical methods.

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Hasegawa group

We have developed spin-polarized scanning tunneling microscopy (SP-STM), and studied magnetic properties of nano-size Co island structures, whose dimension ranges 10-30 nm in lateral size and 1 nm in thickness, formed on Ag(111). From the Moiré intensities observed in STM images, we found two structurally different Co islands on the substrate. Magnetic properties revealed by the SP-STM are also different; one has an out-of-plane magnetization while the other does not. As pristine hcp Co has strong magnetocrystalline anisotropy along the c-axis whereas in fcc Co thin films shape anisotropy prefers in-plane magnetization, we assign the two structures as hcp and fcc Co structures, respectively. We also developed a scanning potentiometry, which enables us to visualize nanoscale spatial distribution of electrical potential under current flow. The potential drops due to finite electrical resistance at grain boundaries were clearly observed in a Au thin film. The method will be applied to two-dimensional surface electron systems that show superconductivity at low temperatures.

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Lippmaa group

This year saw progress on three main fronts: photocatalytic materials, spin filters, and the pyroelectric analysis of ferroelectrics. In the catalyst project, we analyzed the electronic structure of Rh:doped $SrTiO_3$ and showed that the valence state of the Rh dopant can be controlled by a suitable selection of crystal growth parameters. The presence of a mid-gap acceptor state in Rh^{4+}:$SrTiO_3$ was confirmed, explaining the reduced solar energy conversion efficiency. The tunnel junction project culminated with a successful construction of spin-filter tunnel junctions based on ferromagnetic insulator barrier layers. Record-making magnetoresistance ratios were observed in the tunnel junctions. A pyroelectric analysis system was used to analyze the presence of ferroelectricity in Magnetite thin films. A new buffer layer technique was developed for growing perfectly oriented magnetite thin films.

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Division of Physics in Extreme Conditions

Uwatoko group

We report the discovery of a pressure-induced heavy fermion superconductivity in a nonmagnetic orbital ordering state in the cubic compound $PrTi_2Al_{20}$. The strong orbital fluctuations may provide a nonmagnetic glue for Cooper pairing. The results suggest a generic phase diagram hosting unconventional superconductivity on the border of orbital order, paving a new path for further research on novel quantum criticality and superconductivity due to orbital fluctuations. We have performed XAS, NMR, magnetization measurements in relatively large fields and specific heat up to 3 T on heavy fermion $YbCo_2Zn_{20}$ to investigate the possibility that magnetic field gives rise to the delocalization of 4f electrons. Correlations between Yb moments are weak without significant enhancement in the χ(q) spectrum at least in a field, which may explain the lack of magnetic order in $YbCo_2Zn_{20}$ in spite of the existence of distinct local moments. We measured the microwave surface impedances and obtained the superfluid density and flux flow resistivity in single crystals of a phosphor-doped iron-based superconductor $SrFe_2(As_{0.7}P_{0.3})_2$ single crystals (T_C = 25 K). These results indicate the presences of line nodes on at least one band and modulated nodeless gap with deep minimum on the same bands and or other bands. We report a comprehensive high-pressure study on the triple-layer T'-$La_4Ni_3O_8$ with a suite of experimental probes, including structure determination, magnetic, and transport properties up to 50 GPa. The presence of isolated Ni sites with apical oxygen in the T' structure leads to a variable-range-hopping conductivity in the LS phase. The high pressure structural study reveals that a new T^+ structure is stabilized under P>21 GPa. We have performed inelastic neutron scattering measurements in the ferroelectric noncollinear magnetic phase of $CuFe_{0.965}Ga_{0.035}O_2$ under applied uniaxial pressure. The reduction of the spin-lattice coupling, which reflects the partial release of the magnetic frustration due to the nonmagnetic substitution, can also contribute to the emergence of the noncollinear incommensurate magnetic ground state. We have measured the temperature-dependent resistivity of $(TMTTF)_2PF_6$ up to 7 GPa using a turnbuckle-type diamond anvil cell (DAC) and at magnetic fields of up to 5 T. The Ginzburg–Landau coherence lengths for three different axes obtained from this work show that $(TMTTF)_2PF_6$ is an anisotropic three-dimensional superconductor. We have developed a high-pressure and high-field ESR system using the combination of a commercially available SQUID magnetometer up to 5 T and a clamp-type piston cylinder pressure cell up to 1.5 GPa.

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Osada group

(1) We have discussed the mechanism of interlayer surface transport due to helical edge state accompanying the quantum Hall ferromagnetic state in multilayer Dirac fermion systems by employing the tunneling picture. Since this surface state is not topologically protected, it must be diffusive due to spin-inversion scattering. This fact justifies the tunneling picture, in which inter-edge tunneling is regarded as a perturbation. We have concluded that the interlayer tunneling due to the helical surface state is allowed only when the magnetic field is parallel to the side surface of the crystal. (2) We have confirmed the above theoretical prediction by performing the experiment on organic Dirac fermion system α-(BEDT-TTF)$_2$I$_3$. The saturation odf interlayer resistance occurs when the magnetic field was parallel to the stacking direction. Observed features are well explained by the above picture. This agreement indicates the appearance of the QH ferromagnetic phase with the helical edge state in α-(BEDT-TTF)$_2$I$_3$.

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Materials Design and Characterization Laboratory

Y. Ueda group

The materials mainly studied in 2012 are (1) hollandites, (2) A-site ordered manganites, (3) iron-based spin ladder compounds, and (4) low dimensional or frustrated magnetic materials. The main findings in each material group are (1) novel structural distortion accompanied by metal-insulator transition in $K_2Cr_8O_{16}$ and charge order driven by K-vacancy order in $K_xMn_8O_{16}$, (2) Korringa-like relaxation in $YBaMn_2O_6$, (3) block and stripelike magnetism in $BaFe_2Se_3$ and $CsFe_2Se_3$, respectively, and (4) magnetic phases up to 600 T in $ZnCr_2O_4$ and incomplete devil's staircase magnetization curve in $SrCu_2(BO_3)_2$.

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Hiroi group

Various novel compounds are presented, For example, a unique type of frustrated lattice is found in two A-site ordered spinel oxides, $LiGaCr_4O_8$ and $LiInCr_4O_8$. Because of the large size mismatch between Li^+ and Ga^{3+}/In^{3+} ions at the A site, the pyrochlore lattice made up of Cr^{3+} ions carrying spin 3/2 becomes an alternating array of small and large tetrahedra, i.e., a "breathing" pyrochlore lattice. YCr_6Ge_6, comprising a kagome lattice made up of Cr atoms, is a plausible candidate compound for a kagome metal that is expected to exhibit anomalous phenomena such as flat-band ferromagnetism. A unique structural transition in single crystals of the spin-1/2 quasi-kagomé antiferromagnet volborthite, $Cu_3V_2O_7(OH)_2\cdot 2H_2O$, is found, whereby the unpaired electron "switches" from one *d* orbital to another upon cooling, so that it is called the orbital switching transition. Tetrahedral magnetic order and the metal-insulator transition in the pyrochlore lattice of $Cd_2Os_2O_7$ are studied in detail. Rattling and superconducting properties of the cage compound $Ga_xV_2Al_{20}$ are reported.

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Kawashima group

We investigated quantum spin/boson systems and frustrated systems by means of large-scale numerical simulation. We also developed several new numerical techniques. This year we discovered the following facts in particular: (1) super-solid state exists at the commensurate filling in the soft-core Bose-Hubbard model in two and three dimensions, (2) the phase diagram was obtained for hard-core Bose-Hubbard model with dipolar interaction, and (3) the solution of the finite-temperature Gross-Pitaevski equation agree with the Quantum Monte Carlo results even quantitatively.

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Noguchi group

We have studied the structure formation of binary mixtures of two surfactants. We found that various micelles such as bicelles and octopus-like shapes are formed depending on the critical micelle concentration and the ratio of hydrophobic and hydrophilic segments of the molecules. We also studied the dynamics of red blood cells in capillary flow and dynamic-mode correlation in a glass system.

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Materials Synthesis and Characterization group

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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 2012 include that (1) upgrade of the small-angle neutron scattering instrument, SANS-U by introducing a focusing collimation and high-resolution area detector, (2) examination of the theories of rubber elasticity using an ideal polymer network, i.e., Tetra-PEG gels, (3) Rheo-SANS of threadlike micelles, (4) structural characterization of phenolic resin, and so on.

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Yoshizawa group

A systematic study on spin dynamics in two-dimensional transition-metal oxides have been carried out with use of the high resolution chopper spectrometer installed at BL12 in the Material and Life Science Facility, J-PARC. In the highly hole-doped region in the layered nickelate, conventional spin wave excitations change its character to metal-like behavior. Spin dynamics in non-centrosymmetric superconductors $CeRhSi_3$ and $CeIrSi_3$ were also studied.

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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. The first topic of this year is that we developed a calorimetric system for metal hydrides and measured the heat of hydrogen adsorption to palladium and the heat capacities of palladium hydrides. We have clarified that “50 K anomaly”, which has been a long mystery in this material, is due to a glass transition, i.e., freezing of the motion of the hydrogen atoms disordered among the octahedral sites of the palladium fcc lattice. The second topic is that we succeeded to observe magnetic relaxations in a single molecule magnet (SMM) as a clear quasielastic neutron scattering (QENS) for the first time. The third topic is that we found the diffusion of hydrogen molecules in clathrate hydrates stabilized by tetrahydrofuran by means of a QENS technique. Other than these three, we have made some progresses in the studies on porous coordination polymers and ionic liquids.

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Masuda group

The correlation of the higher order of the spin operator has attracted theoretical interest in terms of hidden order in spin disordered state. The challenge is that the experimental probe to identify the correlation is absent. Meanwhile in multiferroic compound that exhibits spontaneous order both in magnetism and dielectricity, the electric polarization is expressed by second order tenors of the spin operators, and the spin nematic operator comes to visible. In this fiscal year our group demonstrated the existence of the spin nematic interaction in an easy-plane type antiferromagnet $Ba_2CoGe_2O_7$ by exploring the magnetic anisotropy and spin dynamics. Combination of neutron scattering and magnetization measurements reveals that the dominant origin of the observed in-plane anisotropy is the antiferro-type interaction of spin nematic operator instead of conventional single-ion anisotropy. The structure of the spontaneous polarization is consistent with the antiferro-nematic order. The introduction of the spin nematic interaction is useful to understand the physics of spin and electric dipole in multiferroic compounds.

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International MegaGauss Science Laboratory

Takeyama group

We have developed a magnetization measurement technique for special use in a vertically aligned single-turn coil, capable of generating ultra-high magnetic fields over 100 T. Compensation ratios of an order as high as 10^{-4} were achieved by self-compensation in parallel twin-pickup coils without using compensation circuit mixing of the signal from an auxiliary coil. High performance cryogenics was employed with a liquid Helium container that was also manufactured to fit the system. The high magnetic field magnetization measurements were applied to a manganite $Bi_{0.5}Ca_{0.5}MnO_3$ at room temperature, and also to a geometrically frustrated spinel oxide, $CdCr_2O4_4$ at very low temperatures. Not only the transition field, but also the absolute value of the magnetization was shown to be evaluated within a 3% degree of accuracy in magnetic fields over 100 T by means of a 14-mm inner-diameter single-turn coil.

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9. *Magneto-Absorption in the α Phase of Solid Oxygen at Megagauss Magnetic Fields: T. Nomura, Y. H. Matsuda, J. L. Her, S. Takeyama, A. Matsuo, K. Kindo and T. C. Kobayashi, J Low Temp Phys **170** (2013) 372-376.

Kindo group

New mono-coil was developed to generate 85.8 T non-destructively. The maximum field beyond 85 T is the world highest field as generated by a non-destructive mono-coil. The coil has an inner bore of 18 mm and the pulse duration is about 4 msec.

1. †*High-Magnetic-Field X-ray Absorption and Magnetic Circular Dichroism Spectroscopy in the Mixed-Valent Compound $YbAgCu_4$: T. Nakamura, Y. H. Matsuda, J.-L. Her, K. Kindo, S. Michimura, T. Inami, M. Mizumaki, N. Kawamura, M. Suzuki, B. Chen, H. Ohta, K. Yoshimura and A. Kotani, J. Phys. Soc. Jpn. **81** (2012) 114702(1-11).

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Tokunaga group

Through the measurements of magnetoresistance up to 65 T, we found field-induced melting of the antiferromagnetic spin order in single crystals of $Fe_{1+y}Te_{1-x}S_x$. Concomitant melting of the orbital order was visually demonstrated through the high-speed polarizing microscopy in pulsed fields.

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Y. Matsuda group

Valence states of rare-earth ions in heavy fermion compounds at high magnetic fields have been investigated. A distinct magnetic field-induced valence transition was observed in $YbAgCu_4$ by the x-ray absorption spectroscopy. The metamagnetism was also found at the valence transition. This findings support the theoretical proposal that $YbAgCu_4$ is located at near the quantum critical point of the valence transition. Moreover, a small magnetic field-induced valence change was detected in a typical Kondo lattice $CeRu_2Si_2$. The observed valence reduction of Ce ion towards 3+ indicates the reduction of the itinerancy of the f-electrons. Apart from the valence states, the microscopic magnetic field-induced spin-crossover phenomenon was observed in a complex molecule Mn III(taa) using the high magnetic field x-ray spectroscopy. The techniques of the x-ray spectroscopy in a pulsed high magnetic field were reported, especially focusing on the portable type capacitor bank and miniature magnets. We have succeeded in making the ultrahigh magnetic field magnetization measurement up to 103 T, and demonstrated the magnetization curves in several magnetic materials.

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Center of Computational Materials Science

Todo group

The main subject of the research in Todo group in FY2012 is as follows: (1) Optimization of transition kernel of Markov chain Monte Carlo. (2) Quantum Monte Carlo level spectroscopy and spin liquid phase in two dimensions. (3) Finite-size scaling with dynamical recovery of isotropy for anisotropic spin systems. (4) Quantum Monte Carlo method for measuring the local Z_2 Berry phase. (5) Deconfined critical phenomena in SU(N) quantum antiferromagnets. We have also developed the ALPS framework for large-scale parallel simulations using quantum Monte Carlo, numerical diagonalization, etc, in order to advance our own research on quantum phase transitions and quantum critical phenomena by eliciting high performance of the modern supercomputers, such as the K computer.

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Laser and Synchrotron Research Center

Suemoto group

The spin reorientation transition in a weak ferromagnet $ErFeO_3$ has been detected by observing the free induction decay signal due to the spin precession motion of the aligned spins, using the terahertz time domain spectroscopy. This demonstrates ultrafast detection of spin orientation with femtojoule pulses which gives practically no thermal disturbance on the sample. The ultrafast hole dynamics, which was hard to access, has been deduced from a comparison of time-resolved luminescence and photoemission data. We propose this type of combined measurements as a new methodology for investigating the hole dynamics in luminescent materials.

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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.

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Takahashi group

We have been studying the structure and phase transition of surfaces and interfaces with diffraction techniques. Bismuth attracts much attention from a viewpoint of topological insulators. It was shown that a single bilayer of Bi(001) on Bi_2Te_3(111) is strongly distorted compared to the bulk Bi, resulting in a large change in the band structure. We have also successfully applied a novel method, analyzing the surface structure as an electron density distribution model-independently, to the structure of a few layers of pentacene on Bi(001).

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Akiyama group

In 2012, we intensively studied physics of short-pulse generation via gain switching of various semiconductor lasers, such as InGaAsP distributed-feedback (DFB) lasers, InGaAs Fabry-Perot (FP) lasers, and InGaN vertical-cavity surface-emitting lasers (VCSELs). We developed quantitative optical spectroscopy and analysis using detailed-balance relations to study non-equilibrium nature of photo-excited carriers in non-doped semiconductor quantum wells, fluorescent radiation thermometry, quantum-wire lasers, and tandem solar cells. We achieved quantitative measurements of color change in spectra of firefly bioluminescence due to modification of enzyme protein structures in site-directed-mutant luciferase and to temperature change. We also accomplished in-situ absorption and fluorescence spectroscopy of firefly oxyluciferin in the wild-type and red-mutant luciferase. Collaborations of theoretical calculations with TD-DFT of electronic states for luciferin and oxyluciferin in water were made to interpret these experiments.

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I. Matsuda group

We have carried out developments and experiments of the advanced spectroscopies using vacuum ultraviolet and soft X-rays.

1) Time-resolved soft X-ray photoemission researches have been performed at high brilliant soft X-ray beamline SPring-8 BL07LSU with the ultrashort pulse laser. By the pump-probe method, relaxation of photoexcited carriers after the surface photovoltage effect at various semiconductor surfaces are traced in real time in the picoseconds to nanoseconds-time scale.

2) The M-edge resonant magneto-optical Kerr effect (RMOKE) of a transition metal film was examined by measurement of the rotating analyzer ellipsometry method and by theoretical simulation based on the resonant scattering theory. The giant Kerr rotation angle over 10 degree at room temperature was observed with vacuum ultraviolet synchrotron radiation at KEK-PB BL-18A. Significant roles of lifetime of the intermediate state during the scattering process and the Fano-resonance were identified. The time-resolved measurement of RMOKE was demonstrated with the SCSS free-electron laser to trace relaxation after the photo-induced demagnetization.

3) The SPring-8 BL07LSU is equipped with a crossed-type long undulator that is designed to generate high-brilliant soft X-rays with various light polarizations. Degrees of linear (horizontal/vertical) and circular (left/right) polarizations were evaluated with soft X-ray polarimeter using the rotating analyzer ellipsometry method.

1. Electronic structure study of ultra thin Ag(111) films modified by Si(111) substrate and by the $\sqrt{3}\times\sqrt{3}$-Ag_2Bi surface: M. Ogawa, P. M. Sheverdyaeva, P. Moras, D. Topwal, A. Harasawa, K. Kobayashi, C. Carbone and I. Matsuda, J. Phys.: Condens. Matter **24** (2012) 115501(1-6).

2. Atomic configuration and phase transition of Pt-induced nanowire on Ge(001) surface studied by scanning tunneling microscopy, reflection high-energy positron diffraction and angle resolved photo-emission spectroscopy: I. Mochizuki, Y. Fukaya, A. Kawasuso, K. Wada, T. Hyodo, K. Yaji, A. Harasawa and I. Matsuda, Phys. Rev. B **85** (2012) 245438(1-6).

3. *Development of soft X-ray time-resolved photoemission spectroscopy systemwith a two-dimensional angle-resolved time-of-flight analyzer at SPring-8 BL07LSU: M. Ogawa, S. Yamamoto, Y. Kousa, F. Nakamura, R. Yukawa, A. Fukushima, A. Harasawa, H. Kondo, Y. Tanaka, A. Kakizaki and I. Matsuda, Rev. Sci. Instrum. **83** (2012) 023109(1-7).

4. Controlling the topology of Fermi surfaces in metal nanofilms: M. Ogawa, A. Gray, P. M. Sheverdyaeva, P. Moras, H. Hong, L. -C. Huang, S. -J. Tang, K. Kobayashi, C. Carbone, T. -C. Chiang and I. Matsuda, Phys. Rev. Lett. **109** (2012) 026802(1-4).

5. Hydrogen-induced surface metallization of $SrTiO_3$(001): M. D'Angelo, R. Yukawa, K. Ozawa, S. Yamamoto, T. Hirahara, S. Hasegawa, M. G. Silly, F. Sirotti and I. Matsuda, Phys. Rev. Lett. **108** (2012) 116802(1-5).

6. *Elucidation of Rh-Induced In-Gap States of Rh:$SrTiO_3$ Visible-Light-Driven Photocatalyst by Soft X-ray Spectroscopy and First-Principles Calculations: S. Kawasaki, K. Akagi, K. Nakatsuji, S. Yamamoto, I. Matsuda, Y. Harada, J. Yoshinobu, F. Komori, R. Takahashi, M. Lippmaa, C. Sakai, H. Niwa, M. Oshima, K. Iwashina and A. Kudo, J. Phys. Chem. C **116** (2012) 24445-24448.

7. Atomic and electronic structures of Si(111)- $\sqrt{21} \times \sqrt{21}$ superstructure: Y. Fukaya, K. Kubo, T. Hirahara, S. Yamazaki, W. H. Choi, H. W. Yeom, S. Hasegawa, A. Kawasuso and I. Matsuda, e-J. Surf. Sci. Nanotechnology **10** (2012) 310-314.

8. Structural analysis of Si(111)- $\sqrt{21} \times \sqrt{21}$-(Ag, Cs) surface by reflection high-energy positron diffraction: Y. Fukaya, I. Matsuda, R. Yukawa and A. Kawasuso, Surface Science **606** (2012) 1918-1921.

9. Time-resolved photoelectron spectroscopies using synchrotron radiation: Past, present, and future: S. Yamamoto and I. Matsuda, J. Phys. Soc. Jpn. **82** (2013) 021003(1-18).

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11. 金属超薄膜内に閉じ込められた電子系のフェルミ面トポロジー制御-量子効果で銀細工: 永村 直佳, 小河 愛実, 松田 巌, 固体物理 **48** (2013) 13-19.

Kobayashi group

We have developed a Dual-comb spectroscopy setup, Multi-GHz repetition-rate mode-locked laser, and precision spectroscopy in VUV.

1. Chirped-pulse direct frequency-comb spectroscopy of two-photon transitions: A. Ozawa and Y. Kobayashi, Phys. Rev. A **86** (2012) 022514(1-6).

2. *Generation of soft x-ray and water window harmonics using a few-cycle, phase-locked, optical parametric chirped-pulse amplifier: N. Ishii, S. Adachi, Y. Nomura, A. Kosuge, Y. Kobayashi, T. Kanai, J. Itatani and S. Watanabe, Opt. Lett. **37** (2012) 97-99.

3. †Vacuum ultraviolet frequency combs generated by a femtosecond enhancement cavity in the visible: B. Bernhardt, A. Ozawa, A. Vernaleken, I. Pupeza, J. Kaster, Y. Kobayashi, R. Holzwarth, E. Fill, F. Krausz, T. W. Hänsch and T. Udem, Opt. Lett. **37** (2012) 503-505.

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6. Comb-Resolved Dual-Comb Spectroscopy Stabilized by Free-Running Continuous-Wave Lasers: N. Kuse, A. Ozawa and Y. Kobayashi, Appl. Phys. Express **5** (2012) 112402(1-3).

7. vuv frequency-comb spectroscopy of atomic xenon: A. Ozawa and Y. Kobayashi, Phys. Rev. A **87** (2013) 022507(1-4).

Itatani group

We have developed a novel BIBO-based intense optical parametric amplifier system that produces 0.55-mJ, 9-fs pulses at 1.6 μm at a repetition rate of 1 kHz with stable carrier-envelope phases. High harmonics are successfully produced with this light source, and we observed CEP-dependent coherent soft x rays that extend to $\backsim$330 eV in photon energies. This is the first observation of CEP-dependent soft-x-ray high harmonics that extend to the water window. The observed high harmonic spectra agree quantitatively well with quantum simulation that suggests the generation of isolated attosecond pulses in soft x rays. This result is an important milestone to extend the attosecond optical science from extremely ultraviolet (EUV) to soft x rays. We have also started to work on high harmonics beamlines for time-and-angle-resolved photoemission spectroscopy (TARPES) and ultrafast magneto-optical Kerr effect (MOKE) spectroscopy for collaborative research on material sciences.

1. *Generation of soft x-ray and water window harmonics using a few-cycle, phase-locked, optical parametric chirped-pulse amplifier: N. Ishii, S. Adachi, Y. Nomura, A. Kosuge, Y. Kobayashi, T. Kanai, J. Itatani and S. Watanabe, Opt. Lett. **37** (2012) 97-99.

2. Sub-two-cycle, carrier-envelope phase-stable, intense optical pulses at 16 μm from a BiB_3O_6optical parametric chirped-pulse amplifier: N. Ishii, K. Kaneshima, K. Kitano, T. Kanai, S. Watanabe and J. Itatani, Opt. Lett. **37** (2012) 4182-4184.

3. 高強度レーザーによる再衝突物理: 板谷 治郎, パリティ **27** (2012) 11-19.

Harada group

1) We have upgraded our ultrahigh resolution soft X-ray emission spectrometer to increase the detection efficiency by implementing a pre-focusing mirror in front of the grating. The upgrade was successful and has improved the performance of the spectrometer by a factor of three without losing energy resolving power.

2) We have studied hydrogen bond property of water using O 1*s* resonant X-ray emission spectroscopy (XES). Spectroscopic evidence of inhomogeneity in liquid water, i.e. tetrahedrally coordinated and highly distorted hydrogen bond picture of water, depends on the character of the pre-edge peak in the O 1*s* X-ray absorption spectrum (XAS). In the recombination emission spectrum at resonant excitation to the pre-edge, we have observed vibrational excitations characteristic of a highly broken hydrogen bond, which strengthen the inhomogeneous picture of liquid water.

3) Determination of oxygen reduction active site in carbon based fuel cell cathode catalyst is a critical issue to systematically improve the catalytic activity. This year we have studied the problem by the following two *in situ* and one *in operando* experiments; i) *In situ* C 1*s*, N 1*s* and O 1*s* XAS, XPS and XES experiments to elucidate O_2 adsorption property of nitrogen doped HOPG as a model of the carbon based catalyst. ii) *In situ* N 1s and O 1*s* XAS and XES to elucidate O_2 adsorption property of highly active carbon based catalyst mixed with Nafion211. iii) In operando XAS and XES to elucidate O_2 adsorption property of carbon based catalyst embedded in a membrane electrolyte assembly (MEA). We have fabricated an original cell for the MEA experiments. The electronic structure of metals in metallo-phthalocyanine derived fuel cell cathode catalysts under operation (gas condition/ bias application) was obtained for the first time. Combining these three experiments we concluded that nitrogen should contribute to the high oxygen reduction activity at high pyrolysis temperatures above 800 °C while at low ($<$ 600 °C) temperatures the residual iron regains oxygen adsorption property through mixture with Nafion and possibly contribute to the oxygen reduction activity.

1. †*Ultrahigh resolution soft x-ray emission spectrometer at BL07LSU in SPring-8: Y. Harada, M. Kobayashi, H. Niwa, Y. Senba, H. Ohashi, T. Tokushima, Y. Horikawa and S. Shin, Rev. Sci. Instrum. **83** (2012) 013116(1-6).

2. *Elucidation of Rh-Induced In-Gap States of Rh:$SrTiO_3$ Visible-Light-Driven Photocatalyst by Soft X-ray Spectroscopy and First-Principles Calculations: S. Kawasaki, K. Akagi, K. Nakatsuji, S. Yamamoto, I. Matsuda, Y. Harada, J. Yoshinobu, F. Komori, R. Takahashi, M. Lippmaa, C. Sakai, H. Niwa, M. Oshima, K. Iwashina and A. Kudo, J. Phys. Chem. C **116** (2012) 24445-24448.

3. * 液体水分子の内殻電子励起ダイナミクスと局所構造: 原田 慈久, 徳島 高, 堀川 裕加, 丹羽 秀治, 木内 久雄, 小林 正起, 尾嶋 正治, 辛 埴, しょうとつ **10** (2013) 14-20.

Joint Use Projects

Jointuse group

1. †*Electrospinning Synthesis of Wire-Structured $LiCoO_2$ for Electrode Materials of High-Power Li-Ion Batteries: Y. Mizuno, E. Hosono, T. Saito, M. Okubo, D. Nishio-Hamane, K. Oh-ishi, T. Kudo and H. Zhou, J. Phys. Chem. C **116** (2012) 10774-10780.

2. †*Hulsite from Sengendera skarn deposit, Miyazaki, Japan: Y. Ogoshi, T. Minakwa and D. Hamane, Japanese Magazine of Mineralogical and Petrological Sciences **41** (2012) 61-66.

† Joint research with outside partners.
* Joint research between groups within ISSP.