The Institute for Solid State Physics The University of Tokyo



Activity Report 2006





ISSP

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Contents	Pages
Preface	1
Research Highlights	2 - 35
Highlights of Joint Research	36 - 51
International Conferences	
and Workshops	52 - 53
ISSP Workshops	54 - 56
Subjects of Joint Research	57 - 101
Publications	102 - 149



ISSP Activity Report 2006

Preface

The ISSP will celebrate its 50th anniversary this fall. It was established in 1957 by the recommendation of the National Council of Japan to set up a new institute for condensed matter physics. The idea was that to catch up the world level of research in the field at that time it was necessary to establish a top class research institute for experimental and theoretical studies of condensed matter and to promote cooperative research throughout the country.



About twenty years after the inauguration the ISSP was recognized as one of the major research institutes in the

community of condensed matter physics and decided to reorganize the institute to promote the project of the "Physics in extreme conditions", which includes ultra-high magnetic fields, laser physics, surface physics, ultra-low temperature physics and very high pressures. After another twenty years, the ISSP completed its relocation in 2000 to the new Kashiwa campus of the University of Tokyo to make further progress and to extend its activities into new fields.

This copy is the Activity Report for the fiscal year 2006 of the research done by the staff of the ISSP and also of the joint research conducted by the ISSP. We are happy to receive any comments on the Report for the possible improvement of the research activities of the ISSP.

At present we are making plans for the events commemorating semicentennial of the ISSP. One is a symposium on the physics of strongly correlated systems, which is one of the major activities of the ISSP. In another symposium the past and present of the ISSP will be reviewed and future directions of the ISSP will be discussed. The fifty year anniversary of the ISSP will be also a good occasion to attract a general audience and to provide them an opportunity to think about the role of materials science in the modern world. The third symposium will be devoted to the general audience including young students. Any suggestions for the events will be most welcome.

June 30, 2007 Kazuo Ueda Director Institute for Solid State Physics The University of Tokyo

Research Highlights

Lattice Preferred Orientation in Post-Perovskite type MgGeO3

Yagi Group

Since the discovery of post-perovskite type MgSiO₃ in 2004 [1, 2], numerous studies have been made to clarify its properties such as stability field, equations of state, elastic property, and plastic property. Because this new phase may explain various strange properties of the layer at the bottom of the lower mantle, a layer called D" layer, which were difficult to explain by any known materials. If the postperovskite phase forms lattice preferred orientation (LPO) in this layer, strong elastic anisotropy observed in D" layer could be explained and various studies have been made to clarify this possibility. Unfortunately the stability field of the post-perovskite type MgSiO₃ is too high to conduct this kind of study and the experiments were made using various analog materials such as MgGeO₃ [3] and CaIrO₃ [4]. Although these two model materials have the same crystal structure, the resulting LPO are quite different and it is difficult at this moment to estimate the behavior of MgSiO₃ in the Earth.

In the present study we have studied the behavior of postperovskite type MgGeO₃ in detail and concluded that the reported LPO in MgGeO₃ [3] is not the deformation texture and cannot be used for the discussion of the D" layer. Experiments were made by squeezing the powdered sample between two diamond anvils and powder X-ray diffraction experiments were made adopting radial diffraction geometry using X-ray transparent gasket [5], as shown in Fig.1.



Fig. 1. Diamond anvil with large side openings developed for the radial diffraction study. Incident X-ray passes through the transparent gasket made of boron and Kapton and irradiates the sample perpendicular to the compression axis of the diamond anvil.



Fig. 2. Unrolled X-ray diffraction profile of the radial diffraction of post-perovskite type MgGeO₃ at 77 GPa. Strong lattice preferred orientation different from the one reported before [ref. 2] is observed.

Orthopyroxene (Opx) type MgGeO₃ was prepared at atmospheric pressure. The powdered starting material was squeezed to 37 GPa at room temperature and converted into perovskite (Pv) structure by heating it using YAG laser. Then further compression was made to 77 GPa at room temperature and then the post-perovskite phase was formed by heating in a uniaxial stress field. Radial diffraction experiments were made at BL13A of the Photon Factory and observed 2D diffraction patterns were recorded on IP detector. The unrolled X-ray diffraction pattern is shown in Fig.2. It is clear from this figure that strong LPO is formed in the post-perovskite phase formed under such condition. However, the observed LPO is quite different from that reported in the previous study [3], which was formed directly from orthopyroxene phase. This result strongly suggests that the LPO in MgGeO₃ so far reported is not the texture representing the deformation fabric but the texture formed by the topotactic relation from the pre phase before the transformation. In order to confirm it, experiments were made to compress post-perovskite phases formed from Opx and from Pv to much higher pressures. The result indicate that the each LPO formed just after the formation remained unchanged even when the pressure is increased to more than 45 GPa after the formation. This is because the sample chamber is too thin to deform during compression. By using diamond gasket technique, we could increase the sample thickness considerably and could see some change in LPO during compression. Further careful studies are needed to clarify the LPO which can be applicable to the discussion of the Earth's deep interior.

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T. Okada, T. Yagi, and K. Niwa

Site-Selective NMR in the Quasi One Dimensional Conductor β-Sr_{0.33}V₂O₅

Takigawa and Y. Ueda Groups

The family of quasi-one-dimensional mixed valent vanadium oxides β - $A_{0.33}$ V₂O₅ (A=Li, Na, Ag, Ca, Sr, and Pb) exhibits a variety of exotic phenomena such as metalinsulator transition, charge ordering, and pressure-induced superconductivity [1], providing fascinating opportunities to study competing electronic orders in one series of compounds. There are three types of one-dimensional V-O network extending along the most conducting *b*-axis (Fig. 1): two types of zigzag chains formed by the V1 and the V3 sites and a two-leg ladder formed by the V2 sites. The ordering of *A* ions doubles the unit cell along the *b*-axis and generates two inequivalent V sites in each of the three 1D structural units denoted as Vna and Vnb (n=1 - 3) in the monoclinic space group $P2_1/a$.

We have performed nuclear magnetic resonance (NMR) experiments on 51 V nuclei in β -Sr_{0.33}V₂O₅, which shows a metal-insulator transition at *T*=170 K (see the article in this report by Y. Ueda's group). We have resolved NMR line from all six V sites in the high temperature metallic phase (Fig. 2) and made site-selective measurements of the Knight



Fig. 1. The crystal structure of β -Sr_{0.33}V₂O₅ with the space group $P2_1/a$ viewed along the (a) *b*-, (b) *a**-, and (c) *c*-directions.







Fig. 3. The Knight shift (K) is plotted against the field direction for all six sites (labeled as A - F) at T=190 K. The angles for the maximum of |K| match precisely with the directions of the shortest V-O bonding in the *ac*-plane, leading to the assignment of the pairs of lines (A, B), (C, D) and (E, F) to (V1a, V1b), (V2a, V2b) and (Va3, V3b) sites, respectively.



Fig. 4. (a): Temperature (*T*) dependence of *K* at various sites normalized by the value at T=300 K. (b) - (d) : *T*- dependences of $1/(T_1T)$. (e): *T*- dependences of $1/(T_1T)$ and $1/(T_1TK^2)$ at the D sites (one of the V2 sites). All measurements were done for the field oriented along one of the "magic angles" shown in the figures, where the quadrupole splitting vanishes.

shift (*K*) (Figs. 3 and 4a) and the nuclear spin-lattice relaxation rate $1/(T_1T)$ (Figs. 4b - 4e) [2].

We found that different sites show contrasting temperature dependences of K and $1/(T_1T)$ (Fig. 4). Thus the magnetic properties are remarkably heterogeneous not only among the three 1D structural units but also among the two V sites in the same unit. In particular, the V2 ladder unit shows pronounced charge disproportionation of *d*-electrons among the two (V2a and V2b) sites as evidenced by the large difference in angular variation of *K* for the C and D sites (Fig. 3). Only one of them (D site) shows strong increase of |K| and $1/(T_1T)$ with decreasing temperature (Fig. 4), suggesting development of ferromagnetic correlation. The strong heterogeneity even in the metallic phase should be a key feature to understand the metal-insulator transition in this family of compounds.

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T. Waki, M. Takigawa, T. Yamauchi, J. Yamaura, H. Ueda, and Y. Ueda.

Angle-Resolved Magnetization Measurements of PrFe4P12: Evidence of a Scalar Order Parameter

Sakakibara Group

Cubic filled skutterudite compounds with the general formula RT₄X₁₂ (R: rare earth; T: Fe, Ru, and Os; X: P, As, and Sb) have been extensively studied recently because they exhibit a number of unusual properties. Among them, one of the fascinating Pr-based skutterudite compounds is PrFe₄P₁₂ which shows an intriguing phase transition at $T_{\rm A}$ =6.5 K. The order parameter (OP) of the phase below T_A (A-phase) is considered to be non-magnetic, because no magnetic superlattice reflection is observed in powder neutron diffraction experiment. In the early stage of the study, the A-phase transition has been thought to be an antiferro-quadrupolar (AFQ) order (alternating alignment of the f orbitals) [1]. Some important features of the A-phase, however, have remained unresolved within the AFQ ordering scenario. First, no significant magnetic anisotropy that could be expected in the AFQ phase develops in the A-phase. Second, no field-induced antiferromagnetic moment perpendicular to applied field has been observed in the NMR experiment [2].

Very recently, a new type of OP that preserves the local cubic symmetry has been proposed to explain the A-phase transition [3, 4]. This order parameter is called the scalar order since the symmetry belongs to the identity representation of the cubic (T_h) point group. A number of unusual properties of the A-phase have been accounted for by assuming a scalar OP. In order to obtain further evidence for the scalar OP, we have studied the field-orientation dependence of the magnetization and the transition temperature in detail [5].

Figure 1 shows the field-orientation dependence of the magnetization M of PrFe₄P₁₂ in the A-phase at T=0.3 K with magnetic fields H rotated in the (1-10) plane. A smooth angular variation of M with weak anisotropy has been observed. The anisotropy is found to diminish as $H \rightarrow 0$. In general, the magnetization can be expanded in odd powers of H as $M = \chi_1 H + \chi_3(h) H^3 + \dots$, where $h = (h_x, h_y, h_z)$ is the unit vector parallel to H. The linear susceptibility χ_1 is isotropic in cubic systems but becomes anisotropic for lower symmetries. The observed weak anisotropy at low fields (see the inset) suggests that the cubic symmetry is preserved in the ordered state. Further evidence of the cubic symmetry is



Fig. 1. Field-angle dependence of the magnetization of $PrFe_4P_{12}$ measured at 0.3 K. The solid line is a fit by the fourth-order cubic invariant term. Inset shows the magnetization curves at low fields.



Fig. 2. Field-angle dependence of the transition temperature of $PrFe_4P_{12}$ measured at 2.8 T. The solid line is a fit by the fourth-order cubic invariant term.

provided by the anisotropic part of M. Within the cubic symmetry, the anisotropic component of the non-linear susceptibility $\chi_3(h)$ obeys the fourth-order cubic invariant $h_4=h_x^4+h_y^4+h_z^4-3/5$ [3]. The solid line in Fig.1 is a fit by the *h*⁴ term, which reproduces the data fairly well.

Figure 2 shows the angular dependence of the transition temperature T_A (*H*) with *H* rotated in the (1-10) plane. A smooth variation is observed over the entire range of the field angle. Here, the solid line again represents the fit by the fourth-order cubic invariant h_4 , in good agreement with the experimental data. These observations strongly suggest that the cubic symmetry is preserved in the A-phase, *i.e.*, the OP is of scalar type.

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Magnetophotocurrent Effect at Low **Temperature in Polymer Solar Cell**

Tajima Group

Polymer solar cells have been intensively studied for several years because of their features such as low cost, low weight, and mechanical flexibility. Today's mainstream device structure is one using a bulk hetero junction, and interpenetrating donor and acceptor materials. Soluble conjugated polymers, such as poly(3-hexylthiophene) (=P3HT) and [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM), which is a soluble derivative of fullerene, are often used as donor and acceptor substances, respectively [1].

From the view point of the organic solid-state science, the bulk-hetero-junction solar cell is an interesting system. This system is an artificial molecular conductor in which carrier density can be controlled by light illumination. No studies have been reported for such systems at low temperatures and high magnetic fields. In this paper, we report on a photocurrent decrease at low temperatures under applied magnetic field. *This is a new phenomenon of organic photovoltaic devices* [2].

We fabricated several devices having a structure of ITO/PEDT:PSS/P3HT:PCBM/Al (see Fig. 1(a)). The thicknesses of the active layer were d = 80 nm and d = 30 nm. The light source was green LED (peak wavelength = 525 nm) chopped at various frequencies.

Figure 1(b) shows the temperature dependences of lightchopped photocurrents at various frequencies for the thin (d = 30 nm) sample. All the curves were measured in one thermal cycle. Although photocurrent decreased on reducing the temperature, the values at 4.2 K were still 1/10-1/5 of those at room temperature. Interestingly, photocurrent exhibits different temperature dependence by changing the chopping frequency. This phenomenon is marked above 100 K.

Figure 2 shows the effect of magnetic field on photocurrent for a thin sample. Thick samples also exhibited a fielddependent photocurrent. By increasing the chopping frequency, and on lowering the temperature, the decrease in photocurrent becomes more prominent. We ascertained that the magnetoresistance in Al and ITO films is negligibly small.

There are several papers reporting on the magnetoresistance and magneto-photocurrent of organic thin films. Frankevich et al. reported the effect of magnetic field on the photocurrent of polymer thin films [3]. In their study, an anomalous increase in photocurrent was observed at a magnetic field below 0.1 T in the temperature range between 130 and 350 K. There are several reports on the anomalous negative magnetoresistance of a particular metal/organic semiconductors/metal junction containing organic molecules [4]. However, we consider that the photocurrent decrease



Fig. 1 (a) A schematic diagram of polymer solar cell. (b) Temperature dependence of photocurrent at various chopping frequencies.



Fig. 2 (a) The effect of magnetic field on photocurrent at 2.0 K. Note that the field dependence differs at two chopping frequencies (20 Hz and 20 kHz). (b) Temperature dependence of the photocurrent below 10 K at B = 0 T and B = 8 T. Chopping frequency is 20 kHz, and the light intensity is 9.25 μ W.

observed in our studies originates from a different mechanism on the basis of the following reasons. First, we observed the photocurrent decrease only at low temperatures, whereas the previously reported phenomena occur at relatively high temperatures. Secondly, we observed a current decrease in the presence of magnetic field, whereas a current increase in the presence of magnetic field was observed in the previous report.

At the present stage, we are considering that the photocurrent decrease found in this study originates from positive magnetoresistance in grains forming bulk-hetero junction. According to literatures, the sizes of P3HT grains in the polymer solar cell increase up to around 8 nm after annealing. The sizes of the PCBM grains are micrometer order. Thus, the grain sizes are comparable to or larger than the magnetic length at 1 T (~26 nm).

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Authors

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Intrinsic Carrier Doping in Antiferromagnetic Supramolecular Copper Complexes: [CuCl₂(Pyra-TTF)]

Mori Group

The synergy or interplay between electrical conductivity, magnetism, permittivity, and optical properties in use of the spin, charge, lattice and molecules degrees of freedom has attracted great interest from the fundamental science to potential applications. Among these applications, the use of organic-inorganic supramolecules has been tried with a view toward the creation of new molecular-based magnetic conductors. Recently, we were the first to synthesize a 3D supramolecular copper(II) complex, [CuCl₂(BP-TTF)] (BP-TTF = bis(pyrazino)tetrathiafulvalene), with a antiferromagnetic interaction between Cu^{II} through organic π -donors, namely π -d interaction [1]. Moreover, we successfully found more conductive supramolecular copper(II) complex [CuCl₂(Pyra-TTF)] (1) by the intrinsic carrier doping from Cu^{II} to donor molecule, Pyra-TTF.

Figure 1 revealed the 3D supramolecular network by the X-ray analysis. The linear chain [-Cu^{II}Cl₂-(Pyra-TTF)-]_n extends along the b axis, where the Cu^{II} atom in a planar geometry is coordinated by two Cl- and two N atoms of Pyra-TTF as ligands [Cu-Cl = 2.23 Å, Cu-N = 2.04 Å]. The Pyra-TTF donors are stacked in a head-to-tail and ring-overbond configuration, where the pyrazino-rings stick out of the donor stacking column. In the *ab* plane, the donors do not form uniform stacking columns, but that two donor units form a honeycomb network. The intermolecular overlap integrals of Pyra-TTF including N atoms are calculated based on the molecular orbitals obtained by WinMOPAC (ver 3.9.0, Fujitsu Limited) to be r = 1.78, p = 0.538 and q =4.64 (×10⁻³). The largest interaction, q, is observed in the diagonal direction, and the second largest one, r, is observed within a unit, so that 2D interactions spread in the *ab* plane. In addition, there exist contacts between donor sheets, C1...Cl⁻¹ (3.35 Å), Cl⁻¹...H1 (2.70 Å) and Cl⁻¹...H2 (2.65 Å). Therefore, this material is the 3D supramolecular copper (II) complex. The temperature dependences of the resistivities of the single crystals of 1 was measured by the ac fourterminal method at 27 Hz with carbon paint contacts. Due to the intrinsic slight carrier doping from Cu^{II} to the coordinated Pyra-TTF, this complex is semiconducting with $\sigma_{\rm RT} = 1.0 \times 10^{-4} \text{ Scm}^{-1} \text{ and } \hat{E}_a = 0.33 \text{ eV}.$

To confirm the intrinsic carrier doping of 1, the temperature dependences of the real (ε ') and imaginary (ε '') parts of the dielectric constants of the single crystal of 1 were measured by impedance analyzer (Agilent 4294A) as shown in Fig. 2. The room temperatures ε ' and ε '' at 1 kHz are 45 and 2240, respectively. The obtained ac conductivity σ =



Fig. 1. Crystal structure of the supramolecular Cu^{II} complex [CuCl₂(Pyra-TTF)] with the 3D framework consisted of the 1D linear coordinated chain [CuCl₂(Pyra-TTF)]_n, 2D donor sheet, and C...Cl⁻, and two kinds of H...Cl⁻ contacts between donor sheets.



Fig. 2. The temperature dependences of real (ε) and imaginary (ε) parts of complex dielectric constants of [CuCl₂(Pyra-TTF)] (1) (// c^*). The obtained ac conductivity $\sigma = 2\pi f \varepsilon_0 \varepsilon$ = 1.25 × 10⁻⁶ Scm⁻¹ (*f*: frequency; ε_0 : vacuum permittivity) and $E_a = 0.39$ eV and the frequency dependence suggests that the carriers of 1 are really doped.



Fig. 3. The temperature dependences of magnetic susceptibility, which follows 2D Heisenberg model from 300 to 4 K with S = 1/2 and 2J = -20 K. The larger antiferromagnetic interaction between Cu^{II} (S = 1/2) spins might be caused not only by 3D interaction but also by the slightly doped carriers in Pyra-TTF donors as ligands.

 $2\pi f \varepsilon_0 \varepsilon'' = 1.25 \times 10^{-6} \text{ Scm}^{-1}$ (f: frequency; ε_0 : vacuum permittivity) and Ea = 0.39 eV and the frequency dependence suggests that the carriers of 1 are really doped. On the other hand, no dielectric response is observed for the insulating [CuCl₂(BP-TTF)]. The temperature dependences of magnetic susceptibilities for 1 was investigated by a Quantum Design MPMS-XL SQUID magnetometer under 10,000 Oe, as shown in Fig. 3. The magnetic susceptibility χ_1 at 300 K after the subtraction of the Pascal diamagnetic contribution is 1.41×10^{-3} emu mol⁻¹ and follows 2D Heisenberg model from 300 to 4 K with S = 1/2 and 2J =-20 K. The larger antiferromagnetic interaction between Cu^{II} (S = 1/2) spins, compared with that of [CuCl₂(BP-TTF)], 2J = -7 K, might be caused not only by 3D interaction but also by the slightly doped carriers in Pyra-TTF donors as lingands. Therefore, our complexes have posed the new material design for molecular magnetic conductors, composed of the conducting donors and the coordinated CuII as a spin source.

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S. Ichikawa, S. Kimura, K. Takahashi, H. Mori, G. Yoshida, Y. Manabe, M. Matsuda, H. Tajima, and J. Yamaura

Unconventional Anomalous Hall Effect Enhanced by a Noncoplanar Spin Texture in the Geometrically Frustrated Kondo Lattice Pr₂Ir₂O₇

Nakatsuji and Sakakibara Groups

Anomalous Hall effect of magnetic conductors has attracted great interest both in the field of fundamental science and applications. Empirically, the Hall resistivity ρ_{xy} is written as $\rho_{xy}=R_0B+4\pi R_sM$ (1), where *B* is the magnetic field and *M* is the magnetization. The first term is the ordinary Hall resistivity. The second term describes the anomalous Hall effect (AHE) whose origin is theoretically attributed to the magnetization and spin-orbit coupling.

Recently, a new mechanism of the AHE, related to the spin chirality under non-coplanar spin configurations has been proposed. A finite spin chirality, which is proportional to a solid angle subtended by spins, induces a finite Berry phase of conduction electrons. This Berry phase acts as a fictitious magnetic field on the conduction electrons and leads to the AHE. Up to now, however, the anomalous Hall effect driven by the spin chirality has been experimentally examined mainly in ferromagnets such as manganites and pyrochlore molybdates. In contrast, a paramagnetic metal with a non-coplanar spin texture is ideal to search for a large Hall effect and to test the validity of spin chirality mechanism. Thus the recently discovered geometrically frustrated Kondo lattice $Pr_2Ir_2O_7$ stands uniquely for such possibility [1].

Here, we report the observation of the divergingly enhanced low *T* Hall resisitvity in the spin-liquidlike regime in Pr₂Ir₂O₇ [2]. In this system, the localized Pr 4*f* moments with <111> Ising anisotropy form a pyrochlore lattice, and provide the non-coplanar spin texture. Ir 5*d*-conduction electrons are only weakly correlated and Pauli paramagnetic. Significantly, because of strong geometrical frustration, Pr₂Ir₂O₇ has no magnetic long-range order down to 120 mK, despite an antiferromagnetic RKKY interaction of $T^*=20$ K between the Pr spins. Instead, the Kondo effect emerges and leads to partial screening of the 4*f* moments below the Kondo temperature of 20 K. On further cooling, the susceptibility exhibits a logarithmically diverging behavior, suggesting that a ferromagnetic coupling eventually develops below 2 K, and the Pr spins form a correlated liquidlike state.

In the spin-liquidlike regime, ρ_{xy} under a field of 0.3 T increases logarithmically on cooling, and reaches a large



Fig. 1. Temperature dependence of the Hall resistivity $\rho_{xy}(T)$ under a field of B=0.3 T along the [111] direction. Inset Left: the Hall coefficient $R_{\rm H}$ vs the susceptibility $4\pi M/B$ under a field of B=0.3 T along the [111] direction. Inset Right: Pyrochlore lattice.

value up to ~ 3 $\mu\Omega$ cm, which is comparable with or larger than typical values for ordinary ferromagnets (see Fig. 1). Furthermore, in the same temperature region, strong anisotropy and non-monotonic dependence on *M* have been observed in ρ_{xy} , which sharply contrasts with the empirical relation given by Eq.(1) (see inset of Fig. 1). Moreover, the field dependence of the Hall resistivity in the low *T* regime does not obey the empirical rule of Eq. (1), but shows strong anisotropy and nonmonotonic *M* dependence.

In terms of the conventional mechanism due to the spinorbit coupling, it is highly difficult to understand the above nontrivial temperature and field dependences of the large Hall effect observed below 2 K. Instead, it is natural to expect the spin-chirality contribution to the Hall effect because the Pr <111> Ising-like spins under fields may well have a sizeable spin chirality due to their noncoplanar spin texture and produce a fictitious magnetic field on the Ir sites through the Kondo coupling. Especially for high fields, we found that the anisotropy of the Hall resistivity can be mainly ascribed to the anisotropy of the fictitious field at the Ir site due to the spin chirality of Pr moments.

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Nonequilibrium Kubo Formula of Finite Conductor Connected to Reservoirs based on Keldysh Formalism

K. Ueda Group

The correlation effects in mesoscopic devices have become a subject of intensive studies. Especially these systems have provided good platform to study the nonequilibrium nature induced by the finite bias voltage. It is well known that in the linear response regime, there is the general relation between fluctuation and dissipation, which is the essence of the Kubo formula. This gives rise to the Nyquist-Johnson relation between conductance and noise power. However concerning the extension of the Kubo formula and the relation between physical quantities in the nonequilibrium situation, there have been few attempts to explore it in mesoscopic systems with interactions.

On the basis of Keldysh formalism we find that the density matrix of mesoscopic systems has a MacLennan-Zubarev form. Using the MacLennan-Zubarev form, it is possible to derive the general formula of differential conductance, which is given by the sum of the well-known currentcurrent correlation function and the non-trivial correlation function between current and the difference of the two particle numbers of reservoirs. In the linear response regime, the differential conductance is given only by the currentcurrent correlation function. Our results show that a naive extension of the fluctuation dissipation theorem into the nonequilibrium case is not possible and requires the additional contribution. We call the generalization the nonequilibrium Kubo formula of a finite conductor attached to reservoirs [1].

Here we stress that our result provides new insight to observable quantities in mesoscopic systems. Let us discuss the nonequilibrium Kubo formula in view of physical quantities. In mesoscopic systems the current-current correlation function expresses noise power at the zero frequency. We would like to propose that the non-trivial correlation function generally gives the formula for shot noise. This fact is checked based on the noninteracting Anderson model, however it is also valid for interacting systems.

Theoretically for noninteracting systems, noise power may be separated into equilibrium noise and shot noise. In this case shot noise can be defined from only noise power. With interaction such a separation cannot be expected in general. In many cases with interaction, it has been considered that shot noise is included in noise power for $eV > k_BT$. Thus the definition of shot noise has been ambiguous in interacting systems. On the basis of the nonequilibrium Kubo formula, it has become possible to define shot noise and give an explicit formula for it. Finally a generalization of the Nyquist-Johnson relation is naturally derived, which we call the nonequilibrium identity between differential conductance, noise power and shot noise. Using the nonequilibrium identity, we may define shot noise experimentally by the difference between observable quantities: noise power and differential conductance.

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Authors T. Fujii

Polarons in Jahn-Teller Crystals: Intrinsic Difference between e_g and t_{2g} Electrons

Takada Group

About a decade ago, Tokura's group made an interesting comparative study of the effective mass m^* between e_g and t_{2g} electrons by measuring the low-temperature electronic specific heat $C_e(T)$ of the manganese oxides La_{1-x}Sr_xMnO₃ as well as the titanium ones La_{1-x}Sr_xTiO₃ [1]. They have found that the t_{2g} (titanium) system has a larger *T*-linear coefficient in $C_e(T)$ than the e_g (manganese) one by more than a factor of two, irrespective of *x*, implying that t_{2g} electrons may be intrinsically much heavier than e_g ones.

Since Mn^{3+} and Ti^{3+} ions are, respectively, considered as typical $E \otimes e$ and $T \otimes t$ Jahn-Teller (JT) centers, we may anticipate that the above experimental result directly reflects an inherent difference in the mathematical structure of JT centers, namely, either $E \otimes e$ or $T \otimes t$. Motivated by this anticipation and as a work subsequent to a previous one of our group for the $E \otimes e$ polarons [2], we have made a comparative study of the polaronic effective mass m^* between the $E \otimes e$ and the $T \otimes t$ JT polarons. A comparison with the conventional Holstein polarons is also made.

If the electron-phonon (el-ph) coupling is weak, it is straightforward to determine the ratio of m^* to the bare band mass *m* by resorting to second-order perturbation calculation, from which we obtain m^*/m as $1+2\alpha$ for both JT polarons with α the nondimensional el-ph coupling constant in its conventional definition. Exactly the same mass ratio can be



Fig. 1. Inverse of the mass enhancement factor, m/m^* , as a function of α for the $T \otimes t$ (solid curve) and the $E \otimes e$ (dashed curve) JT polarons in comparison with the Holstein one (dotted-dashed curve). All the results are obtained in the strong-coupling and/or antiadiabatic region.

reproduced in the Holstein model, if we redefine α in the model in an appropriate way. Thus we see that there is no essential difference in the enhancement of m^* among those three polarons in the small- α limit.

In the limit of $\alpha \rightarrow \infty$, a polaron will be completely localized at a single site, implying $m^*/m = \infty$. For a finite but very large α , the localized polaron will begin to hop between sites, but the hopping in this case is a very rare event. Thus physics connected with such a hopping can be well captured by just considering a two-site system. In fact, by implementing the two-site calculation by exact diagonalization, we can obtain a generic feature of m^*/m in the strong-coupling $(\alpha \rightarrow \infty)$ and/or antiadiabatic (the electron hopping integral *t* much smaller than the local optic phonon energy ω_0) region.

The results of the two-site calculation are shown in Fig. 1 [3]. We note that the Holstein polaron (the dotted-dashed curve) is characterized analytically by the relation of $m/m^* = e^{-2\alpha}$, while the $E \otimes e$ polaron (the dashed curve) by $m/m^* = (\pi \alpha/2)^{1/2} e^{-\alpha}$ [2], indicating the much lighter effective mass of the latter system.

Physically the mass enhancement is brought about by the virtual excitation of local phonons in those model Hamiltonians describing the coupling of an electron with local optic phonons. In the Holstein model no restriction is imposed on exciting multiple phonons, but in the $E \otimes e$ JT model there is a severe restriction due to the existence of a conserved quantity intimately related to the SO(2) rotational symmetry in the pseudospin space representing the twofold degenerate orbitals.

As for the $T \otimes t$ model, no corresponding symmetry exists in the pseudospin space, explaining its difference in m/m^* from the $E \otimes e$ model in Fig. 1. In order to understand its difference from the Holstein model, however, a more detailed analysis is needed. It is well known that the adiabatic potential energy surface for the $T \otimes t$ model contains four equivalent wells for sufficiently large α , but the wells are not isotropic; the vibrational *t*-mode splits into an a_1 mode and two *e*-modes (*i.e.*, $t \rightarrow a_1 \oplus e$). This fact indicates that the $T \otimes t$ model is a combination of the Holstein and the $E \otimes e$ models, explaining the reason why the result of m/m^* in this model (the solid curve in Fig. 1) comes between those of the Holstein and the $E \otimes e$ models.

Our model is too primitive to make a quantitative comparison with experiment, but we find that, at least in the strong-coupling and/or antiadiabatic region, the difference in the behavior of m^* as a function of α can be ascribed to the

presence/absence of rotational symmetry in the pseudospin space representing the internal mathematical structure of each JT center.

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Absence of Aharonov-Bohm Effect in One-Dimensional Bose Liquid

Oshikawa Group

Aharonov-Bohm effect is one of the most striking phenomena in quantum mechanics. It occurs when a quantum mechanical particle is transmitted through a ring interferometer which encloses a magnetic flux, as shown in Fig. 1. In classical mechanics, the path of the particle must take one of the branches. In contrast, according to quantum mechanics, the particle travels both of the branches "at the same time". Namely, the probability amplitude of the particle propagates like a wave, and the contributions through the two branches interfere. This is a famous manifestation of the wave nature of a quantum mechanical particle. The Aharonov-Bohm effect refers to the effect of the enclosed magnetic flux on this interference. In classical mechanics, there should be no effect of the enclosed magnetic flux, as the particle never touches the flux. However, in quantum mechanics, the vector potential rather than the magnetic field is the fundamental quantity, although it is gauge dependent. Even though the enclosed magnetic flux is not touched by the particle, it induces a vector potential which gives rise to a phase difference for the probability amplitude through the two paths. This phase difference is gauge invariant and affects the interference. While being counter-intuitive, the Aharonov-Bohm effect is ubiquitous in quantum mechanics and is related to many important phenomena.

Here we report our recent theoretical work on Bose-Einstein condensate (Bose liquid) confined in one-dimen-



Fig. 1. The ring interferometer is a typical setting for the Aharonov-Bohm effect. The quantum mechanical motion of the particle transmitted through the system exhibits the interference between the probability amplitudes attributed to the two paths. The Aharonov-Bohm effect is the dependence of the interference on the enclosed flux. When the interferometer is filled with Bose liquid, we find that the transport between the two leads is independent of the enclosed (effective) flux, namely the Aharonov-Bohm effect is absent. sional potentials. Nowadays Bose-Einstein condensates can be experimentally realized by cooling of atoms to ultracold temperatures, and are studied vigorously. When the ring interferometer discussed above is filled with the Bose liquid, we found the surprising result that the Aharonov-Bohm effect is absent in the low-energy limit. Namely, the transport through the interferometer becomes completely insensitive to the enclosed (effective) flux [1].

In one dimension, quantum fluctuation is so strong that it destroys the long-range phase coherence. Thus the onedimensional Bose liquid is actually not a true Bose-Einstein condensate. The fluctuation of the local phase can be described by the Tomonaga-Luttinger liquid theory, which is an asymptotically exact effective theory in the low-energy limit. The Tomonaga-Luttinger liquid theory is a universal theory in one dimension, describing also the low-energy physics of one-dimensional electron systems and spin systems. The ring interferometer consists of two Y-shaped junctions of one-dimensional branches. We first studied the Y-junction of three one-dimensional Bose liquids, comparing with our previous analysis on the corresponding electron problem. We found that the strongly coupled junction is stable for a wide range of realistic parameters for Bose liquids, in contrast to the electron systems where an attractive interaction is necessary to stabilize the strongly coupled junction.

We found that a negative density reflection analogous to the Andreev reflection occurs even in the Y-junction of Bose liquids, although the standard Andreev reflection is attributed to the Cooper pair formation of fermions. In the ring interferometer, the Tomonaga-Luttinger liquid can be decomposed into symmetric and antisymmetric combinations of the two branches. The enclosed magnetic flux affects only the antisymmetric mode, which corresponds to the induced persistent current. The transmission between the two leads is only coupled to the symmetric mode, which does not feel the enclosed flux at all. As a consequence, the Aharonov-Bohm effect is absent in the transmission through the Bose liquid in the ring interferometer.

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New Universal Distribution of Spectral-Flow Gaps in the Rashba Spin-Orbit Model

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In recent years, there is a surge of interests in spintronics, *i.e.* to manipulate and to control electron spins in solid state devices by an external electric filed, instead of a magnetic field. Potential advantages include efficient generation of spin polarization inside devices, handy manipulation of individual spins at nano-scales, and desired reduction of heat dissipation during information processing. Since there is no fundamental direct coupling between spin and an electric field, the spin-orbit coupling plays a central role in inducing spin transport. The study of spin transport and, in particular,



Fig. 1. Distributions for level-avoiding gaps.

the spin Hall effect in systems with the spin-orbit coupling has recently attracted much attention. A mechanism for spin transport due to the spin-orbit coupling is called extrinsic or intrinsic, depending on whether disorder (such as impurities or imperfections) is involved or not. In a real sample, there is always disorder. To study possible interplays between intrinsic and extrinsic mechanisms, it is desirable to study them simultaneously in one unified framework, rather than to study them separately and add their contributions. We report such a study of the systems with the spin-orbit coupling using the methods of random matrix ensembles.

A prototype of the intrinsic spin-orbit coupling is the Rashba model in a two-dimensional electron gas. We put the Rashba model with on-site randomness on a cylinder, and include an infinitesimal electric field by turning on slowly a magnetic flux threading the cylinder. The latter is equivalent to imposing a twisted phase in the boundary condition. With a threading flux turned on, the single particle levels will generally avoid, rather than cross. As is shown in Fig.1, our numerical study of level-avoiding gaps in the disordered Rashba model demonstrates that the normalized gap distribution $P_G(\xi)$ is of a universal form, independent of the random strength *W* and the system size *L*. For small gaps it exhibits a linear behavior, while for large gaps it decays exponentially.

In Ref. [1], we suggested a matrix model for the underlying random ensemble, which is verified to exhibit the desired small-gap behavior. Usually one uses the levelspacing statistics as a characteristic of universality in a random system or ensemble. Our results suggest a different type of characterization of random ensembles using gap statistics in spectral flow due to flux threading.

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Advanced First-principles Simulations on Nonadiabatic Dynamics, Electrochemical Reactions, and Phase Transitions

Sugino Group

Elucidating dynamics of electrons and nuclei is an important topic of condensed matter physics and is regarded as a target of recent and future first-principles simulation, although it has thus far focused more attention on the static properties of material. Simulations of the dynamics – transient motion of electrons at femtosecond, adiabatic/nonadiabatic motion of nuclei at picosecond, and fast/slow structural change – have been given more firm theoretical foundations and made more feasible by the development of supercomputers. Therefore computational algorithms are now actively developed to advance the simulations. In this context, we developed algorithms and performed intensive simulations using the supercomputers of ISSP and other centers. Here we report our recent activity.

Near the conical intersection, where two adiabatic potential surfaces cross, the Born-Oppenheimer approximation breaks down even though electron mass is significantly smaller than nuclear mass. The nonadiabatic effect, which thereby gives essentially important contribution to the dynamics, has thus far been solely studied by the accurate configuration interaction (CI) methods. But they become extremely demanding as the size of the system increases. On the other hand, we developed a scheme to compute the nonadiabatic coupling constant using the time-dependent (TD) version of the density functional theory (DFT), which is based on the electron density and thus is potentially applicable to larger systems. Our scheme uses a local approximation within DFT but benchmark calculation with H₃ indicates a CI-level accuracy. The coupled electron-nucleus dynamics has thus made simpler, and intensive applications are now in progress.

At the electrode surfaces, electrons or holes transfer (often rather adiabatically) to nearby molecules triggering an electrochemical reaction. The role of the bias potential applied to the electrode has not been given comprehensive theoretical explanation, in particular for the electrode/solution interface where the water molecules make the reaction extremely easy. Here we used recently developed algorithm called Effective Screening Medium (ESM) [1] to clarify the most standard electrode dynamics, that is, the evolution of hydrogen molecule at the platinum surface. Through intensive simulations, we have found that under a negative bias condition, water molecules form a contact



Fig. 1. A representative configuration of the biased Pt(111)/water interface (left: side view and right: top view). The hydronium ion (yellow circle) is attracted to the negatively charged Pt surface and is weakly bound at the contact water layer that covers the Pt surface with the hydrogen-bond network (right panel). A hydrogen atom is afterwards adsorbed on the surface accepting an electron from the electrode. This is the microscopic aspect of the electrochemical reaction $H_3O^++e^ \rightarrow H(ad)+H_2O$ (Volmer step) in the hydrogen evolution reaction.

water layer at the interface and the double layer away from it. Formation of these structures makes easy the diffusion of the hydronium ions and the electrochemical adsorption process $H_3O^++e^-\rightarrow H(ad)+H_2O$. A general computational scheme for the electrode dynamics has thus been established and is now applied to various electrodes.

Dynamics occur through a very complicated path in some chemical reactions and in most structural phase transitions. To understand them thermodynamically, change in the freeenergy along the reaction path is a very important clue. Existing algorithms to compute the free-energy are not general enough or applicable to extended systems. Here we developed a general algorithm on the basis of the multicanonical ensemble method and benchmarked it using the melting of silicon [2]. The result shows that the applicability of the free-energy-based approach has been greatly extended.

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Phase Transition of Superconducting Wire Network under Fully-Frustrated Condition

Iye Group

The nature of phase transition in artificially structured superconducting systems such as superconducting wire network (SWN) has been the subject of intensive studies both experimental and theoretical. The superconducting phase is described by the complex order parameter $\psi = |\psi| \exp(i\theta)$. Corresponding to the two (amplitude and phase) degrees of freedom, the phase transitions in these systems can occur in two steps. Upon cooling, the amplitude $|\psi|$ of the superconducting order parameter grows first



Fig. 1 (a) Scanning electron microscope image of the Al network decorated with an array of Co dots. (b) Electrode configuration. (c) Cross-sectional view of the segment A-A'. Stray magnetic field emanating from the Co dots creates a staggered flux pattern.



Fig. 2 *top*: The *I*-*V* curves for $\alpha = \beta = 0$. *bottom*: Temperature dependence of the power law exponent *a* of the *I*-*V* characteristics at $\alpha = 1/2$ for several values of β .

around the mean field transition temperature $T_{\rm MF}$ of the superconducting material. Subsequent ordering of the phase θ of the order parameter over the entire system takes place at $T_{\rm c}$ (< $T_{\rm MF}$). It is the latter global phase ordering process in artificially structured superconducting systems that has attracted much interest, as it can be mapped onto the ordering problem of an XY spin system.

In zero magnetic field, the system undergoes the so-called Kosterlitz-Thouless-Berezinskii (KTB) transition, *i.e.* transition via vortex-antivortex pair unbinding. The nature of transition at arbitrary values of frustration parameter α , (α being the average number of flux quanta per plaquette) is not well-understood. The simplest non-trivial case of α =1/2 can be mapped onto the fully frustrated XY (FFXY) model. The vortices tend to arrange themselves in a checkerboard pattern. The system has a double degeneracy associated with the placement of the vortices either in white or black squares of the checkerboard. This leads to occurrence of antiphase domains at finite temperatures, in addition to the vortex pair excitation.

In this work, the nature of transition at $\alpha = 1/2$ is investigated by measureing the *I-V* characteristics, the power-law exponent (*a* of $V \sim I^a$) of which reflects the helicity modulus Γ , or the system's rigidity against phase twist. The effect of staggered field that lifts the double degeneracy is investigated.

We fabricated a superconducting Al wire network decorated by an array of ferromagnetic Co dots as shown in Fig.1. The stray field emanating from the Co dots creates a staggered field of checkerboard pattern whose amplitude β can be controlled by rotating the azimuthal angle of the inplane field and thereby changing the direction of Co magnetization [1].

At zero magnetic field, the system undergoes a KTB transition with the universal jump of a from 1 to 3. For

 α =1/2, T_c is suppressed and the change in *a* at T_c is steeper. These features are consistent with the picture first proposed by Korshnov [2] for the fully frustrated case. According to the model, as T_c is approached from below unbinding of kink and antikink at antiphase domain boundaries effectively destroys the phase correlation across the boundary leading to rapid decrease of helicity modulus.

The effect of non-zero β on the superconducting transition at α =1/2 is to recover T_c and make *a* less rapidly changing at T_c . These features are qualitatively understood by considering the role of β as quenching of the occurrence of antiphase domains At β =1/2 and α =1/2 the canonical KTB transition is restored.

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Berry's Phase Arisen from Spin-Orbit Coupling

Katsumoto Group

When a quantum trips around a loop adiabatically, it acquires a quantum mechanical phase called Berry's phase [1]. When the physical properties of the system (Hamiltonian) are fixed, the phase only depends on the geometry of the loop and hence Berry's phase is also called geometrical phase. The celebrated Aharonov-Bohm (AB) phase is, in a sense, a kind of Berry's phase. (Some of the physicists do not include the AB phase into Berry's phase.) Berry's phase appears in electric transport through quantum mechanical interference effect. The AB oscillation of the resistance in ring-shaped mesoscopic conductors (AB rings) is again a typical example.

When there is a spin-rotation mechanism, such as spinorbit interaction, the interference in the spin part of the wavefunction also plays an important role. In two-dimensional system with strong Rashba-type spin-orbit coupling [2], particles feel effective in-plane magnetic field perpendicular to the momenta. When such a particle is transported along a circle (e.g. by ring shaped boundaries) and the spin-orbit coupling is strong enough, the spin also rotates adiabatically. As the particle goes back to the starting point, the spin has rotated 2π , which is a half rotation for a spinor and thus Berry's phase of π is given to the particle wavefunction. The magnetic field perpendicular to the plane gives vertical component to the spin diminishing the Berry's phase. In the case of an AB ring, this phenomenon results in phase shift in the AB oscillation, which is non-linear for the magnetic field and gives dramatic peak splittings in the Fourier transform spectra [3].

We have observed essentially the same phenomenon in an anti-dot lattice (ADL) system made of two-dimensional hole system (2DHS) at an AlGaAs/GaAs heterointerface [4]. We made circular hollows on the surface of a 2DHS at the square lattice points as shown in the inset of Fig.1. The hollows form repulsion potentials and exclude holes underneath them. Such repulsive dots are called anti-dots. When the lattice constant of an ADL is small enough, the ADL can be viewed as a set of loop orbits encircling anti-dots and



Fig. 1. Fourier spectrum of the oscillatory part in the magnetoresistance of an anti-dot lattice made of a two-dimensional hole system at an AlGaAs/GaAs heterointerface. Clear peak splitting into the structures marked as B, A, A', B' is observed at the main peak. The inset is a scanning electron micrograph of the sample, which has an anti-dot lattice structure.

tunneling connections between them. Then the interference in the loops results in the resistance oscillation called ABtype oscillation.

Figure 1 shows the Fourier spectrum of the oscillatory part of the magnetoresitance. In the main peak structure around the magnetic field that corresponds to single flux quantum per unit cell, clear splitting into three peaks (A, A', B') is observed. Peak A has a shoulder marked as B. These four positions are symmetric to the center indicated as C, where a deep dip is located and just corresponds to single flux quantum per circle with the diameter of the lattice constant. At the second harmonic position (winding number n=2) also a dip indicated as C* appears. Though we do not observe any peak at n=3, a small peak is observed at n=4.

All of these features can be explained by accounting the existence of two different carriers in the 2DHS and the Berry's phase. The results constitute evidence of Berry's phase arising from the spin-orbit coupling and adiabatic rotation of hole spins when they are transformed along circular potentials.

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Reversible Transformation between Spin and Charge Currents Observed in Lateral Ferromagnetic-Nonmagnetic Nanostructures

Otani Group

The basic science for electronic devices aiming at manipulating the spin degree of freedom is spintronics, which provides a possible means to realize advantageous functionalities for spin based recording and information processing. For such functions, the usage of spin current, a flow of spin angular momentum, is indispensable. There is a novel phenomenon where the spin-orbit interaction converts a spin



Fig. 1 (a) Scanning electron microscope (SEM) image of the fabricated spin Hall device together with a schematic illustration of the fabricated device. (b) Schematic spin dependent electrochemical potential map indicating spin accumulation in Cu and Pt induced by the spin injection from the Py pad. Dashed line represents the equilibrium position. (c) Schematic illustration of the charge accumulation process in the Pt wire, where $I_{\rm S}$ and $I_{\rm e}$ denote injected pure spin current and induced charge current, respectively. (d) Spin dependent electrochemical potential map for the charge to spin-current conversion and (e) corresponding schematic illustration.

current into a charge current and vice versa. This is known as the spin Hall effect (SHE), which has recently been demonstrated only by applying optical detection techniques to semiconductor systems such as GaAs at 80 K and Cl doped ZnSe at room temperature, but not yet by the electrical means. In diffusive normal metals, the electrical observation of the inverse SHE has been made by using a nonlocal spin injection in a lateral ferromagnetic-nonmagnetic aluminum structure. However, the observation is limited only to low temperatures because of a small spin-orbit interaction of aluminum. Here we report clear observation [1] of a reversible spin Hall effect (comprising a spin Hall effect and an inverse spin Hall effect), that is the reversible transformation between spin and charge currents both at 77 K and room temperature induced in the platinum wire with a nanoscale spin diffusion length of about 4 nm using a specially designed electrical detection technique combined with the spin-current absorption.

Our device consists of lateral ferromagnetic/nonmagnetic metallic junctions as shown in Fig. 1(a). The device consists of a large permalloy (Py) pad 30 nm in thickness, a Cu cross 100 nm in width and 80 nm in thickness, and a Pt wire 80 nm in width and 4 nm in thickness. The size of the junction between the Py injector and Cu wire, is chosen to be 100 nm × 100 nm to induce an effective spin accumulation in the Cu wire. The distance from the center of the injector to the center of the Pt wire is 400 nm. When the charge current is injected from a ferromagnetic Py pad into a nonmagnetic Cu cross and is drained from one of the top and bottom arms [see inset of Fig. 2(a)], the accumulated spins in the vicinity of the junction induce the diffusive flow of the spin-current

along the Cu wire as can be understood from Fig. 1(b). When the distance between the Py/Cu and Cu/Pt junctions is shorter than the spin diffusion length of about 500 nm, the spin current should be absorbed into the Pt wire from the Cu cross. The injected spin current flows almost perpendicular to the junction plane as in Fig. 1(c). The charge current I_e is generated perpendicular to the spin current Is in the Pt wire via the inverse SHE. One should notice that the inverse transformation is also possible as illustrated in Figs. 1(d) and 1(e).

Figures 2(a) and 2(b) show clear hysteresis both in the charge $\Delta V_C/I$ and spin $\Delta V_S/I$ accumulation signals as a function of the external magnetic field measured at 77 K. Important is that the overall resistance change ΔR_S is exactly the same as ΔR_{SHE} and the same tendency is also observed at RT. This corresponds to the experimental demonstration of the fundamental relations called Onsager reciprocal relations between spin and charge currents. The value of SH conductivity of the Pt wire is evaluated to be 2.4×10^4 (S/m) at RT 10⁴ times larger than the values reported for semiconductor systems and 10 times larger than that for Al. Furthermore, the ratio of the SH conductivity to the electrical conductivity is 3.7×10^{-3} , which is also the largest value reported so far. This result opens up a new possibility to use normal metals with high spin-orbit coupling as spin current sources operating at room temperature for future spintronic applications.

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Fig. 2 (a) The change in Hall resistance $\Delta V_C/I$ due to the inverse Spin Hall effect (SHE) at 77 K. (b) Spin accumulation signal $\Delta V_S/I$ generated by SHE at 77 K. The insets of the figures (a) and (b) show the probe configurations for the measurements.

Self-assembled MnN Superstructure

Komori Group

Self-assembled nanostructures at surfaces have attracted tremendous interest in nanoscience. Among them, the ones with regular spatial array, uniform and well-defined geometric and structural characters, are especially desirable. However, it is still a challenge to prepare such high-quality nanostructures. Recently, we have fabricated self-assembled manganese nitride nano-islands on the Cu(001) surface and proposed a new mechanism of the self-assembly. [1]

As shown in Fig. 1, each island shows a square shape and has a well-defined size of 3 nm \times 3 nm. They are regularly arranged with a periodicity of 3.5 nm and form a two-dimensional square superstructure. The depth of the trench between the islands is the same as the step height of the surface or the height of the island on the superstructure. The nano-islands adopt a 1 \times 1 structure which is oriented in the same way as the fcc Cu(001) substrate. They are reproducibly prepared in three steps in ultrahigh vacuum by Mn deposition, atomic nitrogen exposure and annealing, and stable up to 820 K.

The stoichiometry of the manganese nitride islands is manganese mononitride as determined by in-situ x-ray photoemission spectroscopy in Fig. 2. The MnN film covers



Fig. 1 (a) Large-scale STM image of the self-assembled MnN nanoislands on a wide terrace. Square shaped islands of 3 nm \times 3 nm form a two-dimensional square superstructure. (b) High-resolution STM image. Cross section profile along the blue line is presented in (c). Mn atoms are imaged on the surface, and the island is single atomic-layer high.



Fig. 2 N-1s core-level spectra from a N-saturated $Cu(001)c(2\times 2)$ -N surface and a single-layer manganese nitride covered surface. The coverage of nitrogen on the former surface is 0.5 mono atomic layer (ML). Thus the nitrogen coverage of the latter surface is 0.9 ML.

the whole surface, and is just mono-atomic-layer thick. The bulk MnN crystal has a distorted NaCl structure with a lattice constant 14 % lager than that of the Cu(001) surface [2]. Thus, the nanostructure formation is attributed to strain-relief at the interface. However, different from the conventional stress-domain dominated self-assembly, the shape, size, and periodicity of the MnN islands do not change even when they coexist with the clean Cu(001) surface. This indicates that the self-assembling is driven by a short-range mechanism. The presence of the MnN strips surrounding the MnN island can minimize the strain energy. We propose that MnN island is stabilized by such a simultaneous bilayer organization.

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Regioselective Cycloaddition Reaction of Asymmetric Alkene Molecules with the Asymmetric Dimer on Si(100)c(4×2)

Yoshinobu Group

Potential applications enabled by the chemical modification of Si surfaces include coatings, sensors, molecule-based microelectronics and so forth. The cycloaddition reaction of unsaturated organic molecules with the Si(100) surface is one of the most important families of chemical reactions for the modification of Si surfaces. The asymmetric dimer on the reconstructed Si(100) surface plays an important role, in which a partial charge transfer occurs from a down dimer atom to an up dimer atom. Thus, the chemical nature of Sd and Su is electrophilic and nucleophilic, respectively. In organic chemistry, the regioselective addition reaction of (a)



1. (a) An STM image of 2-methylpropene molecules on Si(100)c(4×2) at 80K. V_s =2.0 V, I_t =0.085 nA. (b) Å contour map of the integrated LDOS on a plane parallel to the surface. (V_s =2.3V, z=11.0Å). The topmost hydrogen atoms of the system locate at z=10.0Å. (c) A structure model of adsorbed 2-methylpropene on Si(100)c(4x2). The large and small open circles represent silicon and budgets according to the function of the system. hydrogen, respectively. The filled circle represents carbon.

asymmetric alkene with an ionic molecule like hydrogen halide is known as "Markovnikov's rule". This reaction undergoes two steps including a carbocation intermediate, and the regioselective product is controlled by the stability of the carbocation intermediate species. In this study, we investigated the adsorption states of 2-methylpropene and propene on Si(100)c(4×2) using low temperature scanning tunneling microscopy [1].

Figure 1(a) shows an unoccupied-state STM image of the 2-methylpropene adsorbed $Si(100)c(4\times 2)$ surface. The protrusions on the Si(100)c(4×2) "bare" surface correspond to Sd sites in the unoccupied state STM image; the large and

small open circles represent the position of the Su and Sd, respectively. The bright protrusions indicated by arrows originate from adsorbed 2-methylpropene molecules. The protrusions of adsorbed 2-methylpropene are obtuse triangleshaped, and the center of the triangle-shaped protrusion is located at the Su position. One of the three corners of the triangle-shaped protrusion points in the dimer bond direction. Previous STM studies attribute the bright protrusions to methyl groups in chemisorbed organic molecules on Si(100). This interpretation is confirmed by the first-principles simulation of STM image in Fig. 1b, which is calculated from local density of states of theoretically optimized adsorption structures (Fig. 1c). Since the observed protrusions are always located on the Su side in the present systems (nearly 100%), we conclude that the alkene carbon atom having two methyl groups is selectively bonded to the nucleophilic Su site while the other carbon atom with two hydrogen atoms is bonded to the electrophilic Sd site.

In order to elucidate the reaction mechanism, we searched for an adsorption structure other than the final disigma state by first-principles calculations. After the structure optimization process starting from various initial configurations with different orientations at the Sd site on Si(100), a stable precursor structure was obtained as shown in Fig. 2. The end carbon atoms without methyl groups are always closer to the Si atom at the Sd site compared with the center carbon atoms with methyl groups in any case. The C=C double bond is elongated to 1.43 Å, while the other C-C bonds in the molecule are slightly shortened from 1.53 Å to 1.51 Å. In addition, the end carbon atom without methyl groups in Fig. 2 shows sp³-like geometrical features; on the other hand, the carbon atoms with methyl groups have sp²like geometrical features. These calculated results indicate that the carbocation-like precursor state plays an important role in these reactions, and Markovnikov's rule works in the present surface reaction.

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Fig. 2. A calculated precursor state of 2-methylpropene on a Si(100) $\sqrt{8} \times \sqrt{8}$ surface. The front view is shown in the circle. The CC and two SiC "bond" lengths are shown in the Å unit.

Element Specific Imaging by Scanning Tunneling Microscope Combined with Synchrotron Radiation Light

Hasegawa Group and Synchrotron Radiation Laboratory

Atomically resolved imaging with a capability of elemental identification is one of the ultimate goals in a development of microscopy. Using scanning tunneling microscopy (STM), which provides us atomically resolved surface images, many attempts have been performed for elementally contrasted images. Most of them, however, merely distinguish one element from others. Here, we report on a new method to obtain element specific imaging using STM. In the method, core electrons of an element are excited with synchrotron radiation (SR) light and emitted electrons are detected with an STM tip. A spatial resolution of the chemical imaging is 14 nm at the present moment and will be improved by using higher brilliance SR beam line [1].

Since STM probes electronic states near the Fermi energy, which are strongly affected by chemical environments, it is difficult to obtain definite "fingerprint" of elements. To obtain elemental information, core electrons, which are characteristic to elements, have to be probed. We have, thus, developed an STM combined with SR so that the STM imaging can be performed under soft-x-ray irradiation, which excites core electrons of specific elements. So far, using the STM, we have taken X-ray absorption spectra (XAS) bearing elemental information of samples by exciting core electrons with the irradiation and detecting emitted electrons with an STM probe tip. In the present study, we successfully obtained element-sensitive images by measuring the photo-induced current during the scanning under photo irradiation whose energy is just above an adsorption edge of the element.

Experiments were carried out at BL-13C at Photon Factory, KEK, Tsukuba. In order to improve detection sensitivity, the light intensity was chopped and the synchronous photo-induced current was measured using a lock-in amplifier. A tungsten tip covered with a glass layer except its apex was used as a probe so that an undesired background component in the photo-induced current is eliminated. A Si substrate having a regular array of Ni dots on it was used as a sample.

XAS spectra measured with an STM tip clearly show peaks due to the Ni L absorption edges (852.7 eV and 869.4 eV). We then took an image of the photo-induced current under the irradiation whose photon energy is just above the adsorption edge (855 eV). The image is presented in Fig. 1 together with simultaneously taken topographic STM image. The site where a Ni dot is found in the topographic image is contrasted bright in the photo-induced current image. As a



Fig. 1. A topographic STM image of Ni-dots fabricated on Si substrate, and simultaneously obtained photo-induced current image. These images were taken under the irradiation of SR light whose photon energy was 855 eV, slightly above the Ni L_3 -edge (852.7 eV).

reference, the same imaging was performed on the same island with a photon energy of 830 eV, far below the Ni absorption edge. Although the Ni dot is clearly visible in the STM image, the photo-induced current at 830 eV does not show any contrast. These results demonstrate possibility of the element specific imaging from a detection of the photo-induced current under the photo irradiation.

Spatial resolution of the element specific imaging was estimated from cross-sectional profiles of the image and found around 14 nm. The observed spatial resolution is better than that of photoemission electron microscopy (PEEM), whose best spatial resolution is currently 22 nm using stateof-the-art instrument with advanced high-brilliance synchrotron radiation light. Our resolution will be also improved by using higher brilliance SR beam line.

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Insulator-metal transition in SrTiO3

Lippmaa Group

Transition metal oxides are well known for their rich electronic and magnetic phase diagrams with many competing phases. In our work, we study carrier density-driven phase transitions in thin oxide layers. By using a field-effect transistor structure (Fig. 1), we can modulate the charge density in a surface layer of a SrTiO₃ single crystal by about 10^{13} cm⁻². This carrier density is sufficient to induce an insulator-metal transition in SrTiO₃. The main advantage of using a transistor to study phase transitions is the ability to continuously tune the carrier density without altering any of the other crystal parameters, such as disorder, which is always created by chemical doping.

Intrinsic SrTiO₃ is an insulator with a band gap of 3.2 eV. The Fermi level is located within about 100 meV of the conduction band bottom, typically within in-gap defect states



Fig. 1. The band structure of $SrTiO_3$ single crystal surface in the vicinity of the conduction band bottom. The Fermi level of the crystal is located within the localized states in the band gap. (Inset) Structure of a $SrTiO_3$ field-effect transistor, showing the insulator layers (yellow) and the channel region where charge is accumulated (blue).



Fig. 2. (a) The channel current of a $SrTiO_3$ FET as a function of gate bias and temperature. At low gate bias, the channel is insulating (violet plots). The channel becomes metallic above a gate bias of 1.3 V (red plots). (b) Scaled sheet resistance of the FET channel under bias. All curves collapse onto a metallic branch and an insulating branch.

that are always present in a crystal, as illustrated in Fig. 1.

Close to room temperature, conductivity is possible due to thermal excitation of carriers from the occupied defect states over the mobility edge. In a transistor structure, a positive gate bias accumulates electrons in the surface layer of the SrTiO₃ crystal, raising the chemical potential closer to the band edge. This results in an drop of the channel sheet resistance, as shown in Fig. 2(a) for low gate bias values.

In this particular device, the chemical potential reached the mobility edge at a gate bias of 1.3 V, which corresponds to a sheet carrier density of 2×10^{12} cm⁻². Above this bias, the device is in a metallic state. When the gate bias is maintained at the critical value, where the chemical potential coincides with the mobility edge, the activation energy for transport vanishes and the sheet resistance is not sensitive to temperature.

It is interesting to note that the transition behavior is, at least qualitatively, similar to the appearance of a metallic state in two-dimensional ultra-clean silicon systems. It is possible to re-plot the sheet resistance data in Fig. 2(a) by assuming a single-parameter scaling relation, $\rho_s(T, n_s) = \rho_s(T/T_0(n_s))$, where T_0 is different for each carrier density value n_s .

When such scaling is performed, all the sheet resistance curves collapse onto two mirror-symmetric branches, one for the insulating side and one for the metallic side of the transition, as shown in Fig. 2(b).

Our work shows that electronic phase transitions can be

successfully induced in a field-effect transistor configuration. The same technique can potentially be used to study a variety of other materials besides SrTiO₃, although achieving a sufficient carrier density modulation remains challenging.

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Magnetization Plateau in Two Dimensional Anti-ferromagnetic Solid ³He on Graphite

Ishimoto Group

³He film adsorbed on graphite surface is one of the most ideal two-dimensional (2D) Fermi systems which can realize not only a Fermi liquid but also a quantum solid just by changing the adsorption density. The transition between liquid and solid is recently considered to be a sort of Mott transition [1]. The 2D solid forms a quantum spin magnet with nuclear spin S = 1/2 on a triangular lattice. Because of the hard-core potential between ³He atoms, the higher order multiple spin exchange (MSE) processes as well as twoparticle exchange play important roles. The exchange of even number of particles is anti-ferromagnetic (AFM), while that of odd ones is ferromagnetic (FM). The competition between them, in addition to the geometrical frustration inherent to a triangular lattice structure, makes the solid ³He film strongly frustrated.

According to recent theoretical studies in the exact diagonalization method for this finite size system, none of long range ordered AFM phases survives even at T = 0 K due to quantum fluctuations. The exchange parameters obtained at high temperatures for the AFM phase (so called a 4/7 phase) suggest a quantum spin liquid (QSL) ground state with a finite excitation gap [2], and a magnetization plateau at around 5 to 6 T. Experimentally the heat capacity and susceptibility measurements indicate a gapless spin liquid ground state [3, 4]. Our magnetization measurement with a



Fig. 1. Temperature dependence of magnetization in the AFM 4/7 phase at various fields. Solid lines are the Brillouin functions where the AFM exchange interaction is taken into account as a molecular field.



Fig. 2. Magnetization curve for the AFM 4/7 phase at 0.68 mK. The vertical axis is normalized by the saturation magnetization.

double-gradient Faraday-type magnetometer shows the existence of magnetization plateau in the AFM 4/7 phase on both ³He and ⁴He mono layer pre-plated graphite. However the exact polarization at the plateau was not yet clear due to a limited S/N ratio.

To clarify the magnitude, we tried to obtain the magnetization curve in the low field side by employing NMR technique where the resonance frequencies are variable in the wide range from a few MHz to 100 MHz in a single experimental run. The paramagnetic first layer solid ³He exhibits a Brillouin function at 3 T in the temperature region down to 0.5 mK, confirming the proper performance of refrigeration and measuring system. Figure 1 shows the magnetization of AFM 4/7 phase on ⁴He mono layer pre-plated graphite as a function of temperature at various magnetic fields. Here the vertical axis is the polarization estimated from the temperature dependence in the high temperature region. The magnetization curve at 0.7 mK is given in Fig. 2, representing a clear plateau at half of saturation magnetization. This is the first step to know the whole magnetization curve and further investigation in the high field region is eagerly desired in spite of many difficulties.

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Torsional Oscillator Experiments of Supersolid State of He and Determination of the Onset Temperature and Characteristic Lengths

Kubota Group

Conventional Superfluidity has been regarded in large as manifestation of unusual properties of Bose Einstein condensation (BEC) with the macroscopic wave function of Bose particles and/or Cooper pairs. The macroscopic coherent system is expressed by a single wave function with an amplitude and a phase. The gradient in the phase implies flow of condensate with v_s . Since v_s is gradient of a scalar field, curl $v_s=0$. Therefore in a singly-connected volume with fixed impermeable boundaries $v_s=0$. In a multiply-connected region there can be a finite circulation with quantization condition. And quantized vortex lines penetrate through the superfluid from rather slow rotation speed Ω_{c1} [1] and a socalled vortex state is formed. Most general situation is vortex lattice formation. Since the first observations of a nonclassical rotational inertia (NCRI) fraction in solid ⁴He [2-6], a vast wave of interest in the supersolid state of matter (see review [7] and references therein) started. An obvious question is what kind of vortex state would be there if supersolid state is similar to conventional superfluids. There are a few quantities necessary to estimate vortex state properties. Namely vortex core size, for example, becomes important to estimate the 2nd critical angular velocity Ω_{c2} , where vortex cores start to overlap each other and finally the 3D superfluidity is destroyed beyond this speed [8].

Another essential, but unsolved experimental problem in the supersolid study has been the determination of the onset temperature T_0 , where this phenomenon starts to appear. T_0 has been reported so far to be about 200 to 300 mK by gradual appearance of NCRI. We have clarified this situation by measuring AC excitation velocity dependence of torsional oscillator, which contains solid He sample. Figure 1 shows the energy dissipation in the solid He sample at 32 bar (upper column) and NCRI fraction (lower column). The upper column data were obtained without any artificial shift of the



Fig. 1. AC velocity dependence of Torsional Oscillator Responses with Solid ⁴He sample at 32 bar pressure. A unique temperature is found near 0.5 K, below which energy dissipation in the solid He sample starts to depend on small changes of AC excitation velocity. We find the nonclassical rotational inertia, or supersolid fraction also starts to appear and change depending on the AC velocity below the same temperature. The inset shows energy dissipation data with a clear peak for another solid ⁴He sample. It is remarkable to see the small AC velocity influences very much of the supersolid property. On the other hand we find that AC velocity dependence of the energy dissipation per supersolid fraction has a maximum near 200 μ m/s, which seems to have different origin as apparent critical velocity which characterizes only NCRI decreasing velocity. We can argue provisionally that what we found is real bulk energy dissipation critical velocity whereas the NCRI critical velocity is a characteristic velocity which indicates fluctuation of NCRI component.

data at all and we can see clearly that all the data start to change depending on small AV velocity below a certain temperature near 0.5 K. We can associate the same temperature for the change of NCRI as well as seen in the lower column. These observations give us opportunity to determine the onset temperature T_0 for sure, thereby confirming that supersolid transition is not associated with possible liquid phase which might have remained in solid samples [7].

By utilizing above information of T_0 we could analyze the supersolid fraction temperature dependence and proceed to evaluate Josophson's length, for the first time. Vortex core size is expected to be the same order as Josephson's length [8] and we can use it to estimate vortex state parameters. We obtained two sets of length scale, which may reflect the appearance of vortex fluid state and the original supersolid state as recently discussed by P.W. Anderson [9].

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Angle-Dependent Magnetotransport Study in an Organic Conductor: Test for Unconventional CDW State

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The unconventional charge density wave (UCDW) state, where quasi-particles have nodal dispersion, has been recently discussed in organic conductors α -(BEDT-TTF)₂XHg(SCN)₄ (X=K, Rb, Tl). In contrast to the case of unconventional superconductivity, the zero-gap quasi-particles could contribute to the transport phenomena. It has been discussed that the anomalous angle-dependent magnetoresistance oscillation (AMRO) appears resulting from the Landau quantization of quasi-particle spectrum. The unexplained AMRO similar to the Lebed resonance, which is observed in those organic salts, has been the main experimental basis of the UCDW model.

In order to check the possibility of UCDW, we have experimentally and theoretically investigated the Lebedresonance-like AMRO observed in the CDW phase. Detailed stereographic measurements of interlayer magnetoresistance in X=K salts have revealed that the amplitude of AMRO is modulated in the different way from the conventional Lebed resonance (Fig.1). We have found that the modulation pattern quantitatively coincides with Kajita oscillations of the remaining cylindrical Fermi surface (FS), which does not contribute to the CDW formation. This fact strongly suggests that the observed AMRO originates not from the quantized quasi-particles but from the cylindrical FS.

Next, to confirm if the observed AMRO could be explained as an effect of the cylindrical Fermi surface, we have evaluated interlayer conductivity using the quantum tunneling picture, which considers only the lowest-order contribution of single interlayer tunneling between neighbouring two layers. We have successfully reproduced the observed features, that is, the coexistence of Lebed resonances and Kajita oscillations as shown in Fig.2. This effect is the AMRO of the magnetic breakdown system with the CDW potential.

In conclusion, the anomalous AMRO observed in the CDW phase in α -(BEDT-TTF)₂XHg(SCN)₄ originates from the cylindrical FS. The present result is the negative one against the UCDW model.

Authors

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Fig. 1. Interlayer resistance of α -(BEDT-TTF)₂KHg(SCN)₄ as a function of magnetic field orientations. The amplitude of Lebedresonance-like AMROs (dashed line) are modulated by Kajita oscillations (horizontal solid lines).





Magnetization Measurements by Using Turnbuckle Type Diamond Anvil Cell –Magnetic Behavior of TmTe–

Uwatoko Group

The Tm monochalcogenide series TmX (X=S, Se, Te) has the NaCl structure and is among the mysterious strongly correlated electron systems. Many aspects of TmSe have been investigated including the valence fluctuation between Tm³⁺ and Tm²⁺, the Kondo effect, antiferromagnetic ordering ($T_N = 3.4 \text{ K}$ [1]), and the metal-insulator transition. It was shown that the behavior of the electrical resistance was not that of a typical metal, but displayed a Kondo lattice-like behavior. The Te analog, TmTe, exhibits anomalous high pressure electrical resistance behavior above 2 GPa [2] similar to the Kondo-like behavior seen in TmSe. To provide a more complete understanding of this behavior, we have performed single crystal magnetization measurements to 6.1 GPa.

The magnetization was measured by a Quantum Design SQUID magnetometer. Pressure was generated in a novel turnbuckle diamond anvil cell (DAC) with a diameter and height of 6.4 mm and 7.4 mm, respectively. Figure 1 shows a cross section of the CuBe alloy DAC. The sample (TmTe single crystal) had dimensions of 45 μ m × 180 μ m × 180 μ m and was placed into the 400 micron hole of a NiCrAl alloy gasket together with ruby chips and the pressure-transmitting medium, glycerin. The pressure was calibrated by measuring the shift of the ruby fluorescence line, R1, at 80 K. In conventional high-pressure-magnetic experiments, the large cell background makes it difficult to observe the sample signal. To overcome this, a large sample was used and the cell background was subtracted. The NiCrAl alloy gasket was not aged, since the aged material has a large magnetic moment if not handled correctly.

The field cooled (1000 Oe) temperature dependent magnetization traces are shown in Fig. 2. It was found that the temperature dependent magnetic susceptibility for 0 GPa and 2.56 GPa showed paramagnetic behavior with no discernible distinction between the two traces for temperatures between 2 K and 80 K. Also, the magnetic field dependent magnetization curves have been performed at 2.0 K. The 1.0 GPa curve shows paramagnetic behavior. A small hysteresis and saturation in magnetism were observed at 4.7 GPa indicative of ferromagnetism. From the saturation of the magnetic



Fig. 1. Diamond anvil cell, designed for magnetic measurement by a SQUID magnetometer and pressures up to 10 GPa.



Fig. 2. 1000 Oe field cooled temperature dependences of the magnetization. On upper figure, with increasing pressure, the temperature of the transition decreases. The magnetic susceptibility for 3.6 GPa shows a paramagnet-ferromagnet transition at $T_{\rm C}$ = 12.2 K, which was ascertained by differentiating the magnetic susceptibility. $T_{\rm C}$ decreases with increasing pressure, disappearing above 6.1 GPa within the accessible temperature range. We note that the moment increases with decreasing temperature below 8.9 K at 3.6 GPa. This same phenomenon is seen in the 5.4 GPa data. The origin is unknown.

moment, the effective magneton numbers at 4.7 GPa and 5.4 GPa are 1.85 μ B/f.u. and 1.62 μ B/f.u., respectively. That is bigger than the value 1.2 μ B/f.u. at 5.4 GPa found by neutron diffraction experiments [3]. The moment is smaller than that for Tm²⁺ (4 $\mu_B/f.u.$) and Tm³⁺ (7 $\mu_B/f.u.$). Ferromagnetic behavior was not observed at 6.1 GPa. Electrical resistivity measurements have been performed under high pressure [2]. At 5.7 GPa the resistivity showed an abrupt decrease which was believed to be due to a structural phase transition. We observed that the ferromagnetic phase disappeared between 5.4 GPa and 5.85 GPa. From these results, below 2.56 GPa, TmTe displays paramagnetic behavior. Above 3.6 GPa, a ferromagnetic phase suddenly appears. With increasing pressure, the transition temperature decreases. By X-ray absorption spectroscopy in the partial fluorescence yield mode, the valence of Tm in TmTe is mostly divalent for 0-3 GPa and 2.45 for 3-4.5 GPa [4]. This may be the cause of ferromagnetic phase appearing in this pressure range.

Magnetization measurements were performed under extremely high pressure using a diamond anvil cell. The ambient and 2.56 GPa data showed similar behavior for temperatures between 2 K and 80 K. At 3.6 GPa, the temperature dependence of magnetization shows the onset of a paramagnetism-ferromagnetism transition at $T_{\rm C}$ = 12.2 K. On increasing the pressure, $T_{\rm C}$ decreases. Above 6.1 GPa, the transition disappears. We propose that a change in the valence value of Tm plays an important role in this behavior.



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Lissajous-like Motion of the Wave-packets in a Two-dimensional Adiabatic Potential Well

Suemoto Group

Recently, we have been studying the nuclear wave packet dynamics in solids by using femtosecond time-resolved luminescence spectroscopy. The wave-packet movie has been taken for the self-trapped excitons (STE) in brominebridged platinum complexes (Pt-Br) [1]. Selective sensitivity of this method to the excited state has been demonstrated for F-centers in alkali-halides [2].

Here, we present an evidence for Lissajous-like motion of the wave-packet on a two-dimensional potential surface observed in cooled Pt-Br [3]. The red curves in Fig. 1 are the time evolution of the luminescence intensity at 0.95 eV (top) and 0.85 eV (bottom). The process we are observing is shown in the inset of Fig. 2. The system is excited (upward arrow) from the ground state (blue bowl) to the excited one (red bowl). The wave-packet starts from the upper part of the red bowl and makes an oscillatory motion. We observe the luminescence at certain points on the red bowl, shown by a downward arrow. When the wave-packet passes on this



Fig. 1. Time evolutions of the luminescence intensity at 0.95 and 0.85 eV. Red and blue curves correspond to experiment and calculation, respectively. The large peaks at t=0 in the experimental curves are tentatively assigned to the radiation before self-trapping. The numbers (1-6) correspond to those in Fig.2.



Fig. 2. Potential energy surfaces of the self-trapped exciton for the ground state (blue) and the excited state (red). The yellow disk is the wave-packet and the green curve is its trajectory. The inset shows the optical transitions between the two-dimensional potential surfaces.

point, the luminescence flashes. The fine oscillatory features with a period of ca. 300 fs in the red curves in Fig. 1 correspond to this flashing. In addition to this main oscillation, we can notice a complicated behavior, *i.e.*, the intensity is modulated by a slower oscillation component at 0.95 eV. On the other hand, at 0.85 eV, the oscillation persists for about 2.5 ps without this modulation and stops suddenly.

These complicated waveforms are understood in terms of a model assuming a two-dimensional damped oscillation on the excited state potential well. The wave-packet (yellow disk) moves on a trajectory shown by a green curve (see Fig. 2). The tilted black line shows the position where the luminescence photon energy is 0.95 eV. Crossing or approaching of the wave-packet to this black line, shown by orange ovals, bring about increase of the luminescence intensity. The simulated time evolutions are shown in Fig. 1 by blue curves. The numbers (1-6) indicated correspond each other in Fig. 1 and Fig. 2. The important features are well reproduced in terms of this simple model. More elaborate calculation based on resonant secondary radiation theory coupled with an interaction mode description of the wave-packet was performed [3].

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Highly Advanced Coherent Soft X-ray Generation Down to 7.9 nm Due to High Harmonic Based on Novel Scheme of Laser-ablation Controlled Plasma

Kuroda Group

Since the time when first ruby laser was demonstrated, future development of x-ray laser remains as one of a dream of laser physicists and almost all researchers engaged in laser



Fig. 1. HHG spectra from tin and vanadium laser ablation irradiated by femetosecond laser pulse. The intensity of the 17th harmonics was 20 times higher than those of its neighbors. The conversion efficiency of the 17th harmonic was measured to be about 1.1×10^{-4} , and this output energy of 1.1 μ J was obtained from the pump laser energy of 10 mJ.

applications involving biomedical applications and basic medicine. High-order harmonic generation (HHG), which occurs as a result of the interaction of strong laser fields with atoms, ions, or molecules, is one of most promising short wavelength unique source of coherent soft x-rays with distinct advantages of high brightness, good beam quality, and femtosecond, or even attosecond duration. The demonstration of the HHG applications such as the control of electron dynamic in molecules, nonlinear optical phenomena in the XUV region, the photoelectron spectroscopy and ultrafast dynamics of multiphoton-induced photoelectron emission has recently been reported. Thus the HHG have been recognized as very attractive tools. For the application of the HHG emission, it is important to increase the conversion efficiency of this process. In addition, it is necessary to extend the cutoff energy of the harmonic. From these objectives, we developed the high intensity harmonic generation at short wavelength region (high photon energy region).

For the high intensity HHG, we report first demonstration of the single harmonic enhancement by using multiphoton resonance with the strong oscillator strength ions at the wavelength of 47 and 37 nm [1, 2]. Figure 1 shows the spectra of HHG from the laser-ablated tin plume. A strong 17^{th} order harmonic at a wavelength of 46.76 nm was observed using tin laser ablation plume, as shown in Fig. 1. The conversion efficiency of the 17^{th} harmonic was measured to be about 1.1×10^{-4} , and this output energy of $1.1 \,\mu$ J was obtained from the pump laser energy of 10 mJ. Figure 2 shows the HHG spectra from the laser-ablated tin



Fig. 2. HHG spectra from tin laser ablation for pump laser with central wavelength of (a) 795 nm, (b) 782 nm, and (c) 778 nm. The ordinate of spectra (b) and (c) are 6 times increased. The intensity of the 17th harmonic gradually decreased as the central wavelength of laser become shorter.

with three different laser wavelengths of 795 nm, 782nm, and 775 nm. In Fig. 3(a), one sees that the intensity of the 17th harmonic using 795 nm wavelength pump dominates the harmonic spectrum. The intensity of the 17th harmonic is 20 times higher than that of other harmonics. However, in Fig 3(b), the intensity of the 17th harmonic using 782 nm wavelength pump is decreased, and is almost same as that of other harmonics. In Fig. 3(c), the intensity of the 17th harmonic with 775 nm wavelength pump is further decreased. In the case, the 17th harmonic intensity is weaker than that of the 13th and 11th harmonics. The above results show that the intensity of the 17th harmonic gradually decreased as the wavelength of the pump laser become shorter. As mentions above, the origin of this enhancement is attributed to resonance with strong radiative transition of the Sn II ion, produced within the laser-ablated plume. For the laser-ablation Sb, we successfully demonstrated the intensity enhancement of a single high-order harmonic at a wavelength of 37.67 nm. The conversion efficiency of this harmonic was 2.5×10^{-5} and the output energy was 0.3 µJ. Such an approach can pave the way toward a considerable enhancement of a single harmonic in the short-wavelength range using the appropriate target materials.

On the other hand, for the extension of cutoff energy in high harmonic (HH), we demonstrated first observation of HH by using the *doubly charged ion* in the laser-ablation plume via produced by pre-plasma. By using the laserablation V plume, we observed the 71^{st} harmonic at the photon energy of 110 eV. Very recent result of the HH cutoff has successfully extended up to 150 eV by using the laserablation Mn plume [4]. Thus the HH from the lowly charged ion have obtained the quite novel results, especially in their outstanding high efficiencies and attractive physics in related ion and atomic plasma laser physics.

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Laser Controlled Nanostructures Formation on Wide Band-gap Semiconductors Using Two-beam Femtosecond Laser Pulses

Kuroda Group

Advanced laser nano-fabrication has great potential applications in optoelectronic and information technology, and it has been studied intensively in recent several years. Many kinds of nanostructures, such as nanogratings, nanowires and three-dimensional photonic crystals were fabricated by femtosecond laser ablation method. Wide band-gap semiconductors have excellent optical, thermal and



Fig. 1. The dependence of An on laser wavelengths λ . Here A is the nanograting period, n is refractive index. The solid line represents $\lambda/2$.

mechanical properties, and they attract much attention in optoelectronic industry. We developed a pump-probe method, and studied the formation of nanostructures on wide band-gap semiconductors induced by femtosecond laser pulses.

We report nanogratings with periods of 150 and 80 nm formed on the surface of 6H-SiC crystals irradiated by the ppolarized 800 nm and the s-polarized 400 nm femtosecond lasers, respectively. When both of the two collinear laser beams focus simultaneously on the sample surface, nanoparticles formed on the whole ablation area, and they array in parallel lines. If we change the laser wavelength with OPA (optical parametric amplifier) system, nanogratings with periods in the range of 80-250 nm were fabricated [1, 2].

We report a simple route of growing ZnSe nanowires on ZnSe crystal surface [3, 4]. Uniform ZnSe nanowires are observed on the ablation crater irradiated by femtosecond lasers in air, while other part of the sample surface is not damaged. The nanowire growth rate is about 5 µm/s, it is higher than that fabricated by chemical vapor deposition method by a factor of 10⁴. The nanowire length and diameter can be controlled by varying laser pulse energy and pulse number. If the crystal wafer is horizontally dipped in pure water, needle-like nanowires are also found in the ablation craters. Our discovery opens a rapid and simple way to grow nanowires on the designed micro-patterns.

Besides the formation of nanostructures, we also studied the ultrafast dynamic in dielectrics and multilayers irradiated by femtosecond laser [5, 6]. We found avalanche ionization plays an important role in the processes of the generation of conduction band electrons and sample damage.

Nanofabrication on dielectrics, semiconductors and metals using soft x-ray laser is a hot research topic. In our group, we have developed a novel scheme of laser-ablation



Fig. 2. SEM images of the ablation craters and nanowires. The laser parameters are: (a) and (b), 800 nm, 120 fs, linear polarization, 1 mW 500 pulses; (c) and (d), 400 nm, 120 fs, linear polarization, 10 mW, defocus for 3 mm, 500 pulses.

controlled plasma and realized highly advanced coherent soft x-ray due to high harmonic generation. In the near future, we will extend the formation of nanostructures and related physics to a new stage. By utilizing the soft x-ray coherent lights developed in our group, we will study the high resolution microfabrication using ultra-short x-ray pulses.

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Photoluminescence of Single GaInAsP/InP Quantum Wires Formed by Etching-Regrowth Techniques

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Recent advances in the top-down nano-technology techniques of GaInAsP/InP quantum-film (Q-Film) epitaxial growth, electron-beam lithography, dry etching, and regrowth, provide excellent controllability in fabricating various nano-structure devices operating at 1.5 µm wavelength, such as low-threshold quantum-wire (Q-Wire) lasers for telecommunication applications [1]. However, no optical measurement has ever been reported on a single Q-Wire fabricated by this method, due to material damages introduced during the etching-regrowth processes and also to



Ga_{0.25}In_{0.75}As_{0.50}P_{0.50} (-0.15% TS Barrier 12nm)

Ga_{0.22}In_{0.78}As_{0.81}P_{0.19} (1% CS Well 6nm)

OCL: Optical Confinement Layer TS: Tensile-Strained CS: Compressively Strained

Fig. 1. Schematic of a Q-Wire sample. Excitation energy was 1.26 eV, where the surrounding InP was transparent at the energy.



Fig. 2. Normalized PL spectra for Q-Wires with various W (as denoted) and a Q-Film at 4 K. Peak energy of the Q-Film is represented by a vertical broken line. Three series of peaks denoted as P1, P2, and P3, were clearly observed. Intense PL at 1.1 eV is due to OCLs.

low sensitivity of spectroscopic systems around $1.5 \,\mu\text{m}$ wavelength. In this work, we measured and analyzed photoluminescence (PL) spectra at 4 K of etching-regrowth single GaInAsP/InP Q-Wires with lateral widths down to 6 nm and lateral quantum confinement energies up to 90 meV.

Figure 1 shows schematic of an etching-regrowth single GaInAsP/InP Q-Wire sample. The sample was grown by organometallic vapor-phase-epitaxy (OMVPE) technique in two steps. In the first growth, an undoped strain-compensated single 6 nm-thick GaInAsP Q-film structure sandwiched by optical confinement layers (OCLs), or cladding layers, was grown on a n⁺-InP substrate. After the growth, Ti metal patterns of 0.91 mm-long isolated wires with various widths in the nm scale were formed with electron-beam lithography and a lift-off process. Then, CH₄/H₂ reactive ion etching and additional slight wet chemical etching for cleaning were carried out to form single Q-Wires. The Q-Wires were buried in intrinsic InP regrown in the second growth step. The final Q-Wire widths W = 6 - 40 nm were characterized by cross-sectional scanning electron microscope measurements.

We performed micro-PL scanning measurements on single Q-Wires with a spatial resolution of about 1.5 μ m limited by the excitation spot size. Figure 2 shows observed normalized PL spectra at 4 K of the Q-Wires with various W. The spectrum of the narrowest Q-Wire (W = 6 nm) is shown at the top, and spectra of wider Q-Wires are sequentially shown below. As a reference, a PL spectrum of the as-grown Q-Film in the same original wafer was measured and shown at the bottom. As the width W of Q-wires decreases, PL peaks denoted as P1, P2, and P3 showed systematic energy blueshifts from the PL peak energy of the Q-Film. Since P2 existed in all Q-Wires and converges to the Q-Film peak, this was ascribed to a recombination between electrons and holes in the ground subbands in a Q-Wire. The energy blueshift represented lateral quantum confinement, which is as large as 90 meV at the W = 6 nm spectrum. While P3 is ascribed to one of the excited subbands in the Q-Wires, the origin of P1 is still unknown. All the peaks P1, P2, and P3 were experimentally ascribed to the Q-Wires, because scanning measurements across the Q-Wires have shown that they emerged only when the spot overlapped with the Q-Wires. In the scanning measurements along the Q-Wires, we observed uniform spectra over 30 μ m, which revealed high homogeneity of the Q-Wire sizes along the Q-Wires.

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Development of Copper-Coated Insertion-Device Vacuum Chamber

Nakamura Group

Insertion devices (IDs) can provide highly brilliant light for SR users and enable the users to do state-of-the-art experiments. On the other hand, ID vacuum chambers can generate large long-range wakefields by interaction with a multi-bunch electron beam and cause resistive-wall coupledbunch instabilities of the beam, because they have a very small aperture and are made of stainless steel in order to obtain high mechanical strength in spite of thin thickness and to reduce eddy current effects in the magnetic gap or phase changes. For reduction of the resistive-wall wakefields, we proposed that the inner surfaces of the ID stainless-steel vacuum chambers should be coated with a highly conductive metal such as copper [1]. However, effects of copper coating on the ultra-high vacuum conditions had not been investigated. Therefore we studied and developed a copper-coated chamber usable under the ultra-high vacuum conditions [2].

A developed copper-coated chamber was produced as a test chamber for measurement of the outgassing rate by



Fig. 1. Photograph of test copper-coated chamber.



Fig. 2. Setup for measurement of the outgassing rate of the coppercoated chamber.

plating all the inner surface of a stainless steel chamber (1.4 m in length and 152 mm in diameter) with copper. In this plating procedure, the stainless steel chamber was put into the copper sulfate solution and then dipped for about 3 hours. The thickness of copper coating was about 150 µm, which is sufficiently thick for suppressing the wakefields significantly. Figure 1 shows the photograph of the test Cucoated chamber. For comparison, a test stainless steel (SS) chamber usable under the ultra-high vacuum conditions was also produced. It had the same length and diameter. The orifice method was applied to measuring the outgassing rate of the test Cu-coated and SS chambers. Figure 2 shows the set-up for measurement of the outgassing rate. This system consists of the test Cu-coated chamber (or SS chamber), two B-A gauges (BAG), an orifice (\$\$ 5mm) between them, a Qmass analyser, a metal valve, two molecular pumps (TMP), two gate valves in the upstream of the TMPs and two cold cathode gauges (CCG). First we opened the metal valve and pumped the entire chamber during baking out and then closed the metal valve after the baking out. The gas from the inner surface of the chamber passes through the orifice and is pumped out by the TMP1 as shown by the arrows of Fig.2. The outgassing rate q from the chamber under the equilibrium condition is described by $q=C(P_1-P_2)/A$, where P_1 and P_2 are the measured pressures by BAG1 and BAG2, C is the conductance of orifice and A is the area of the inner surface of the chamber.

Table 1 shows the results of the outgassing rate measurement under the ultra-high vacuum. For each test chamber, the total baking time was 200 hours at 300°C and then the test chambers were cooled down for 48 hours at 24°C. The measured outgassing rate of the SS chamber was consistent with the other experimental data [3] and the measured outgassing rate of the Cu-coated chamber was smaller than that of the SS chamber. In addition, the measured mass spectrum of the Cu-coated chamber was almost the same as that of the test SS chamber. It was concluded from the measurement results that the developed copper-coated chamber was usable under the ultra-high vacuum conditions. Recently the same copper coating was adopted for a new ID vacuum chamber of KEK-PF BL-16 to keep the beam stable.

chamber	outgassing rate	
	[Pa m ³ s ⁻¹ m ⁻²] (N ₂ , 24°C)	
Cu-coated	(0.90±0.06)×10 ⁻¹⁰	
SS	$(1.36\pm0.06)\times10^{-10}$	

Table 1. Results of outgassing rate measurement

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Hyper Gels with High Mechanical Properties: Clay-Polymer Nanocomposite Gels

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In general, gels are made of a small fraction of polymer network component and a large portion of solvent. Hence, gels are very eco-friendly material and are served in various applications. Though high toughness and mechanical strength in gels have been anticipated, conventional gels are very brittle and fragile. Haraguchi et al. reported a novel clay-polymer nanocomposite gels (NC gels) consisting of synthetic hectorite and poly(N-isopropyl acrylamide) (PNIPA) [1]. This "nanocomposite hydrogel" (after their notation) has excellent physical properties, such as tough mechanical properties, large deformability, large swelling ratio, rapid shrinking capability, and high transparency [2]. For example, NC gels are much tougher than conventional PNIPA hydrogels (organic gels; OR gels) by a factor of 20 -50 in deformability and of 300 - 1000 in toughness. The most interesting fact is that the improved properties are obtained by simply replacing N,N'-methylenebis(acrylamide) (BIS) (cross-linker of usual chemical gels) with clay before polymerization of PNIPA. Note that a simple mixture of PNIPA and clay in water does not behave like NC gels. In order to elucidate the origin of the superiority in the mechanical properties of NC gels, we carried out small-angle neutron scattering (SANS) experiments and discussed the deformation mechanism.

SANS experiments were carried out at SANS-U, ISSP with 7.0 Å-neutron beam. Figure 1 shows the two-dimensional scattering intensity patterns of uniaxially stretched NC gel [3, 4]. Here, q is the magnitude of the scattering vector. The NIPA and clay concentrations were 0.04 M and 700 mM, respectively. The molecular weight of PNIPA is expected to be more than 2.0×10^3 kg/mol. The SANS patterns clearly show that the scattering profiles become anisotropic by stretching. Even in the case of contrast-



Fig. 1. Two-dimensional SANS patterns for NC4 (SDD = 8m); upper (in D₂O solvent) and lower (in contrast matched solvent). The stretching direction is horizontal and λ denotes the stretching ratio.

(a) NC gel



Fig. 2. Deformation model of (a) NC gels and (b) OR gels. PNIPA chains in NC gels are effectively bridging clay platelets and the molecular weight is rather high compared with those of OR gels.

matched sample, where the scattering contrast between water and clay is matched by tuning hydrogen/deuterium ratio, the scattering patterns accompanying two lobes in the stretching direction are clearly observed. Since clay platelets are supposed to be "invisible", this two-lobe pattern must be ascribed to an anisotropic scattering from deformed polymer chains. At the high-*q* region, on the other hand, the SANS pattern stretched in the perpendicular direction is interpreted to be due to elongated polymer chains along the parallel direction. Hence, by combining ordinary and contrastmatched SANS, we could differentiate the two contributions, *i.e.*, clay platelet orientation and polymer chain stretching, and conjectured the following mechanism.

Figure 2 shows the difference in the structure of NC gels and OR gels [3]. First of all, the "spatial" inter-cross-link distance of a NC gel, *i.e.*, the inter-platelet distance is much longer (of the order of 400 Å) than the inter-cross-link distance of OR gels (of the order of 10 to 20 Å for the case of 700 mM PNIPA gels with 5% cross-links) [5]. Secondly, the "topological" chain lengths $\langle R^2 \rangle^{1/2}$, are also very different, where R is the end-to-end distance of PNIPA chains between cross-links and < > denotes ensemble averaging. That is, the molecular weight of PNIPA chains bridging neighboring clay platelets is much larger than that of subchains between cross-links for OR gels. The former could be of the order of 10^5 to 10^6 [6], while the latter is of the order of 10^1 to 10^2 (based on the ratio of NIPA and BIS). Thirdly, the number of "effective" cross-links, which are capable of transmitting the external load, is much larger in NC gels than in OR gels. This is because most of PNIPA chains in NC gels are anchored to clay platelets and play a role as active chains, while most of PNIPA chains in OR gels are separated by cross-linkers to short chains and merely form clusters with a high density of cross-linking. These facts directly result in the high elongation ratio and high strength at break of NC gels. Upon stretching, clay platelets become oriented with their surface normal parallel to the stretching direction. It is concluded that the extraordinarily good mechanical properties are ascribed to "plane crosslinks" with long polymer chains between platelets and high density of "force sustainable" polymer chains.

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Localized Mode in the β-Pyrochlore Superconductors AOs₂O₆

Hirota and Hiroi Groups

Yonezawa et al. [1, 2, 3, 4] have recently discovered a new series of superconductors AOs₂O₆ (A=K, Rb,Cs). The superconducting transition temperature falls with increasing the ionic radius of the alkali metal ion: $T_c = 9.6, 6.3, 3.3$ K for A = K, Rb, Cs. While the origin of their superconductivity was studied intensively, other interesting features were also revealed: a localized mode of the alkali ion in all the three compounds and a mysterious first-order phase transition in KOs_2O_6 below T_c . An alkali ion is located in an oversized cage made of Os and O (See Fig. 1). Due to weak bonding between the alkali ion and the cage, it is expected that the alkali ion vibrates independently of the cage. Hiroi et al. [5] pointed out the existence of this independent vibration (localized mode) macroscopically through specific heat measurements. The first-order phase transition was also discovered by Hiroi et al., and a small structural change has been found. Now we speculate that anharmonicity of the localized mode plays a significant role in the phase transition.

In order to identify the localized mode *microscopically* and to evaluate its anharmonicity, we performed neutron scattering experiments. Inelastic neutron scattering (INS) experiments and neutron powder diffraction (NPD) experiments were performed on AOs_2O_6 (*A*=K, Rb, Cs) powder samples using the ISSP triple-axis spectrometer PONTA and the IMR powder diffractometer HERMES installed at the JRR-3 reactor in JAEA. In INS experiments, we successfully observed a low-lying peak around 6 meV, which indicates that the localized mode exists in all the three compounds (See Fig. 1). The values of the energy of the mode are



Fig. 1. Inelastic neutron spectra from AOs_2O_6 . (open circle: A = K, closed square: A = Rb, open triangle: A = Cs) Inset: crystal structure of the β -pyrochlore superconductors AOs_2O_6 .



Temperature (K)

Fig. 2. Temperature dependence of the atomic displacement parameters of AOs_2O_6 . The solid curves are the calculated values of harmonic oscillators with the energy determined from INS experiments.

consistent with those determined in the previous specific heat experiments in $RbOs_2O_6$ and $CsOs_2O_6$ while disagrees in KOs_2O_6 . This discrepancy suggests strong anharmonicity of the mode in KOs_2O_6 . In NPD experiments, we found that atomic displacements of Rb and Cs ions are harmonic but that of the K ion is quite anharmonic (See Fig. 2). Thus we revealed the existence of the localized mode in all the three compounds microscopically and the strong anharmonicity of the vibration of the K ion.

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Proton Dynamics in Highly Proton-conductive Copper Rubeanate

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It is a great and urgent subject in current material science to develop high-performance proton conductors for hydrogen storage and fuel cell technology. Copper rubeanate (CuC₂N₂S₂H₂) is a porous coordination polymer exhibiting high proton conductivity at room temperature. Nagao *et al.* [1] have revealed that this compound absorbs water molecules depending on relative humidity (RH) and its proton conductivity is enhanced by four decades. The conductivity at RH=100% is 0.01 S/cm which is as large as that of the best proton conductor known before. Figure 1 shows the schematic crystal structure of copper rubeanate including water molecules inside the pores.

We have measured inelastic and quasielastic neutron scattering data from copper rubeanate (CR) and its hydrated sample (CR-3.6H₂O) to investigate the mechanism of proton conduction, especially the role of water molecules. Neutron scattering method is quite effective for studying dynamics of



Fig. 1. Schematic structure of copper rubeanate including water molecules in pores.

protons since the incoherent neutron scattering cross section of H atom (81.67) is much larger than those of other atoms (*e.g.*, C: 0, N: 0.49, O: 0, S: 0.01, Cu: 0.52). We used a high resolution pulse cold neutron spectrometer AGNES [2], which is installed by ISSP at C3-1 guide of JRR-3 (Tokai, JAEA).

Figure 2 shows the temperature dependence of elastic scattering intensities, reflecting mean square displacement of H atoms, of both dry and hydrated CR. The data of bulk H₂O are also plotted for comparison. The data of dry CR and ice lie on each straight line as predicted by the harmonic oscillator model. The intensity of ice decreased drastically at the fusion temperature ($T_{\text{fus}} = 273.15$ K) and the slope of the data became steeper above $T_{\rm fus}$ due to diffusive motions of water molecules. We have found that CR hydrate exhibits excess decrease of elastic intensity (bending of the straight line) and corresponding quasielastic neutron scattering (broadening of the elastic peak) above 200 K. Each quasielastic spectrum was fitted well by the combination of Lorentz and a Gauss functions. The momentum transfer (Q)dependence of elastic incoherent structure factors (EISF) revealed that water molecules are not moving around the crystal but localized in the pores. The Q dependence of the HWHM of Lorentzian was reproduced by the jump diffusion model as well as in bulk water. In Fig. 3, the logarithmic self-diffusion constants D of bulk H₂O and CR hydrate are plotted against inverse temperatures. It is of interest that the activation energy of CR hydrate is much smaller than that of bulk H₂O. This result suggests that protons of CR hydrate



Fig. 2. Temperature dependence of logarithmic elastic scattering intensities of dry and hydrated copper rubeanates and bulk H₂O. The inset shows the quasielastic spectrum observed at 340 K and the result of fitting by Lorentz (blue curve) and Gauss functions.



Fig. 3. Arrhenius plots of self-diffusion constants of copper rubeanate hydrate and bulk water. The activation energies were obtained from the slopes of the fitted straight lines.

are carried by the Grotthuss mechanism (combination of H_3O^+ reorientation and H^+ hopping along O-H...O hydrogen bonds). We are now measuring the neutron scattering data of a related compound CuC₂N₂S₂(C₂H₄OH)₂ and its hydrate sample. This compound has more hydrophobic pore walls than CR and so will provide further information on the role of water molecules in the proton conducting process.

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Representation Analysis Approach for Magnetic Structure Determination

Sato Group

A salient feature of neutrons is their magnetic-dipole interaction with spins in magnetic materials; magnetic scattering of neutrons is a direct consequence of the interaction. Hence, in principle spin structure may be determined by analyzing neutron scattering data. A key for successful spin-structure determination is selection of initial spin-structure models. However, to date this is largely based on experiences using a so-called trial-and-error method, which makes spin-structure analysis non-trivial for non-specialists. Therefore, a systematic method for the construction of the initial models is highly desired.

Magnetic representation analysis, introduced by Izyumov *et al.* [1], provides such a systematic way for the initial structure selection. In this method, an initial model may be obtained by a linear combination of a selected number of irreducible magnetic representations of the paramagnetic phase; coefficients in the linear combination automatically become fitting parameters, instead of spin directions S_x , S_y , and S_z with *a priori* constraints as assumed in usual spin structure analysis.

Although the mathematical theory of the magnetic representation analysis was constructed more than 20 years ago, it has not been widely used because of its cumbersome mathematics. Several computer programs have been developed to obtain magnetic representations recently. However, they are not seamlessly combined with the diffraction data analysis. Hence, to make a full use of the magnetic representation analysis, we have started developing the representation analysis and profile fitting code. At the present moment, we have successfully developed the representation analysis code called mbase, and profile fitting code masa-tof, which is specific to the time-of-flight diffractometer LAM-80D installed at the KENS spallation neutron source, KEK, Japan. As an example of their applications, in what follows we will present our recent spin-structure analysis on the ternary rare-earth compound CeB₂C₂.

Space group of CeB₂C₂ is known to be tetragonal $P_{4/mbm}$; the Ce atoms occupy the 2(*a*) site with the site symmetry C_{4h} [2]. This compound shows a magnetic order at $T_N = 7.3$ K, where an incommensurate spin structure was inferred [3]. The details of the spin structure have not been elucidated to date. We have performed the neutron powder diffraction experiment using LAM-80D, in a hope that this will confirm our approach to the diffraction analysis.

Shown in Fig. 1 are the resulting neutron diffraction patterns in a temperature range spanning T_N . As seen in the figure, new Bragg peaks appear in the diffractogram below T_N . The magnetic reflection positions were successfully indexed by using the magnetic modulation vector of $q = (\delta, \delta, \delta')$ where $\delta = 0.167(3)$ and $\delta'=0.114(3)$.

The magnetic representation vectors for the Ce sites were obtained for the given q vector (and symmetrically equivalents); of total six representation vectors, we try to find the simplest combination that reproduces the observed diffraction pattern. As a result, a good coincidence between the observed and calculated profiles is found for a combination of IR2:1 + IR2:2. The basis vectors for the IR2:1 are $a_{2:1,1} = (1, 0, 0)$ for the Ce (0, 0, 0) site, and $a_{2:1,2} = (0, e^{2\pi i \delta}, 0)$ for the Ce(1/2, 1/2, 0) site, whereas those for the IR2:2 are $a_{2:2,1} = (0,1,0)$ and $a_{2:2,2} = (e^{2\pi i \delta}, 0, 0)$. The resulting spin structure



Fig. 1. Neutron diffraction patterns in $Ce^{11}B_2C_2$ obtained using LAM-80D. In the lower part of the figure, the peak positions and intensities are schematically shown for several spin structures defined by linear combinations of the irreducible representations IR*n*:*m*.

is sinusoidally modulated with the spin polarization along [110] direction. It should be emphasized that the validity of the spin structure is not quantitatively claimed but only qualitative comparison can provide evidence for the presently proposed structure; as seen in the lower part of the figure, only IR2:1+IR2:2 structure can reproduce the observed peak positions.

As demonstrated here, the representation analysis is a very powerful tool for the spin structure determination. We are now working on the application of the presently developed codes for reactor based diffraction data.

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Multiple Ground State Competition under Pressure in β-Sr_{0.33}V₂O₅

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β-vanadium bronzes are a member of vanadium bronzes with a chemical formula $A_x V_2 O_5$ in which the stoichiometric composition is x = 0.33 (1/3). There are six compounds of β - $A_{0.33}V_2O_5$ (A = Li⁺, Na⁺, Ag⁺, Ca²⁺, Sr²⁺, Pb²⁺). The monoclinic crystal structure consists of a characteristic V₂O₅-framework and A-cations. The A-cations are located in the tunnels formed by the V₂O₅-framework and occupy only one of the two nearest-neighboring A-sites within the a-c plane in a zigzag-type ordered manner along the *b*-axis. The V₂O₅-framework is formed by sharing the corners and edges of (V1)O₆, (V2)O₆ and (V3)O₅ polyhedra, where V1, V2 and V3 are the three crystallographically independent vanadium atoms. Each polyhedron forms three kinds of infinite chain along the *b*-axis (the conductive direction in these quasi-onedimensional conductors). Among six compounds, all compounds except Pb-compound exhibit metal-insulator (MI) transitions with the nature of charge order (CO) at independent temperatures, accompanied by the formation of superstructure with a common 6-fold periodicity ($q_b = 1/6$) along the b-axis. This 6-fold periodicity comes from 2-fold by A-cation order multiplied by 3-fold by CO. The ground state of insulating phase is Néel ordered state in A+compounds and spin-gapped state in A^{2+} -compounds except Pauli-paramagnetic Pb-compound. We have revealed that the CO transitions are suppressed by hydrostatic high-pressure and that the superconductivity appears in A^+ -compounds, while in A^{2+} -compounds no superconductivity is observed.

Very recently, magnetic susceptibility (χ) and resistivity (ρ) of β -Sr_{0.33}V₂O₅ were successfully measured under high pressure [1]. Under ambient pressure, β -Sr_{0.33}V₂O₅ exhibits a sudden increase of χ at CO transition temperature ($T_{\rm CO}$ = 150 K) and below the temperature it shows a low dimensional spin gap behavior with a broad maximum around 50 K. With increasing pressure, $T_{\rm CO}$ is gradually lowered and some hump in χ -*T* curve appears around 50 K and 0.5 GPa in addition to the jump at $T_{\rm CO}$. This hump grows as pressure further increases and then χ -T curves around 0.6 GPa clearly shows three transitions around at $T_{\rm CO}$ = 140 K, $T_{\rm d}$ = 65 K



Fig. 1. Pressure-temperature (*P*-*T*) phase diagram of β -Sr_{0.33}V₂O₅. The filled and open symbols are derived from magnetic susceptibility and resistivity measurements, respectively. The colored stars represent the superperiodicity with $q_b = 1/2$ (2*b*), 1/6 (6*b*) and 1/10 (10*b*) observed in x-ray oscillation photographs.

and $T_i = 40$ K. These are regarded as the transitions between the different electronic states with various lattice modulations such as $q_b = 1/6$ and 1/10, which have been confirmed by x-ray diffraction under high-pressure. With increasing pressure, both $T_{\rm CO}$ and $T_{\rm i}$ decrease, while $T_{\rm d}$ increases and merges with $T_{\rm CO}$ around 1.1 GPa. In addition to these three anomalies in the CO phase, a kink both in χ -T and ρ -T curves was observed at around $T_{\rm m} = 140$ K in the charge disordered (CDO) phase (the metallic phase). The transition might be an antiferromagnetic (AF) ordering, although there is no direct evidence. Very similar behaviors of χ -T curves under pressure have also been observed in β -Ca_{0.33}V₂O₅. Furthermore the χ -T curve of β -Pb_{0.33}V₂O₅ shows an anomaly around 60 K above 0.4 GPa, which suggests any magnetic transition. Thus we can give a speculation for the absence of superconductivity in A^{2+} -compounds, namely AF order is realized before the system falls into superconductivity. The obtained P-T phase diagram (Fig. 1) displays the competition among multiple ground states under pressure.

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Search for Missing Link – Binary Vanadium Oxide V4O9–

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Vanadium atom takes various oxidation states, which gives us rich variety of binary vanadium oxides. In addition to V₂O₃, VO₂ and V₂O₅ containing vanadium atoms with integer oxidation states, many mixed-valent compounds, Magnéli phases V_nO_{2n-1} (n = 3-9) between V₂O₃ and VO₂, and V_nO_{2n+1} (n = 3, 6) between VO₂ and V₂O₅, are reported. Most of them have been intensively studied in the viewpoint of correlated electron systems, especially of their metal-



Fig. 1. Temperature dependence of magnetic susceptibility (χ) for V₄O₉. $\chi(T)$ is obtained by subtracting Curie term and temperature independent term from the raw data (χ_{raw}). The inset shows a crystal structural model of V₄O₉.

insulator transitions.

In V_nO_{2n+1}, V₄O₉ with a mixed-valence state of V⁴⁺/V⁵⁺ = 1, has been missing. We have succeeded in synthesizing single phase powder samples, solving its crystal structure, and elucidating physical properties of V₄O₉. Though V₄O₉ is easily decomposed during its synthetic process, we found a proper condition to synthesize single phase V₄O₉ samples from V₂O₅ by reduction controlled precisely. By combining x-ray, neutron, and electron diffractions, a crystal structure model was built and refined. Different from other binary vanadium oxides, the crystal structure of V₄O₉ has VO₄ tetrahedra, indicating the existence of definite V⁵⁺ sites. Remaining highly distorted VO₆ octahedra containing V⁴⁺ form ladders, which are bridged by VO₄ tetrahedra, as shown in Fig. 1.

Reflecting the crystal structure, V4O9 is an insulator. Magnetic susceptibility behavior is of low dimensional magnetism as shown in Fig. 1, suggesting the existence of spin gap $\Delta / k_{\rm B} \sim 70$ K. Magnetization curve measured up to 70 T also indicates a spin gap ground state. Magnetic excitation energy measured by inelastic neutron scattering using powder samples is about 8 meV, which is consistent with that from magnetic measurements. Momentum dependence of inelastic neutron scattering data reveals a weakly interacting spin dimer system.

Compared with a famous spin gap system $(VO)_2P_2O_7$, V_4O_9 has a similar structure with VO_4 tetrahedra instead of PO₄ tetrahedra in $(VO)_2P_2O_7$. We can expect almost the same physics behind these two systems.

Authors

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Metal-Insulator Transition in Ca_{1-x}Na_xIrO₃ with Post-Perovskite Structure

Y. Ueda and Yagi Groups

CaIrO₃ with *Cmcm* orthorhombic symmetry captures great geophysical interests, since the isostructural MgSiO₃ is considered to be the main constituent of the D" region in the lowermost mantle [1, 2]. The crystal structure (right inset of

Fig. 1), which is now called the post-perovskite structure, comprises IrO₃ layers and eightfold-coordinated Ca²⁺ ions in between. Within each plane, IrO₆ octahedra are connected with each other by sharing edges and corners along the *a*-and *c*-axes, respectively, giving rise to a rectangular lattice of Ir⁴⁺ ions with the t_{2g}^5 electron configuration (S = 1/2). This electronic state well resembles that of the parent compounds of high- T_c cuprates. Turning our attention to this similarity, we introduced mobile carriers into CaIrO₃ by substituting Na⁺ for Ca²⁺ [3].

Figure 1 represents the electronic phase diagram for $Ca_{1-x}Na_xIrO_3$ ($0 \le x \le 0.37$) within a temperature-Na content (T-x) plane. The parent compound (x = 0) is a Mott insulator, which undergoes the antiferromagnetic transition at 115 K. Upon carrier doping, the resistivity (ρ) gradually decreases until a metallic state characterized by $d\rho/dT < 0$ is eventually achieved at $x \sim 0.30$. At this composition, the magnetic ground state simultaneously changes from the antiferromagnetic to paramagnetic one. The absence of an intervening antiferromagnetic-metallic phase between the antiferromagnetic-insulating and paramagnetic-metallic phase markedly contrasts with many metal-insulator transition (MIT) systems in three dimensions; instead, this resembles high- T_c cuprates with a quasi-two-dimensional lattice. However, superconductivity is indiscernible in the vicinity of the MIT; this is likely related to the fact that a doped carrier forms a spin triplet state in the present system (left inset of Fig. 1), in contrast to the Zhang-Rice singlet in high- T_c cuprates.

The metallic state on the verge of the MIT shows some intriguing properties that cannot be understood in the framework of the Fermi liquid theory. The $\rho \propto T^{\alpha}$ ($\alpha = 1.2$) relation indicates presence of the antiferromagnetic fluctuation; this actually manifests itself in the strongly *T*-dependent susceptibility. The ρ shows $\ln T$ divergence at low *T*s probably owing to the weak localization effect in two dimensions. The *T* dependence of the orbital magnetoresistance is so strong that Kohler's rule does not hold in the present system. It should be noted that these three properties are also observed in high-*T*_c cuprates, and their origins are still under debate. The observation of these phenomena in Ca_{1-x}Na_xIrO₃ offers a solid foundation to unravel anomalous electronic properties in the normal state of high-*T*_c cuprates.



Fig. 1. The electronic phase diagram for Ca_{1-x}Na_xIrO₃ ($0 \le x \le 0.37$) within a temperature-Na content (*T*-*x*) plane. Left inset: Schematic view of the electron configuration among the t_{2g} orbitals. One doped carrier denoted by an open circle forms a triplet (high-spin) state with the preexisting carrier. Right inset: Post-perovskite structure composed of IrO₃ layers, and Ca²⁺ and Na⁺ ions in between.

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Computational Experiment on Field-Cooled Magnetization Processes in Ising Spin-Glass Model

Takayama Group

One of most fundamental problems yet unsettled in spinglass (SG) studies concerns the stability of the equilibrium SG phase under a finite magnetic field h. By experiments at an early stage of the study, the field-cooled magnetization (FCM), the magnetization measured while a spin glass is slowly cooled under h, was observed to exhibit cusp-like behavior at a certain temperature T_{cusp} . This FCM behavior is roughly consistent with the prediction of the SG meanfield theory with T_{cusp} being interpreted as the critical temperature of the SG transition under h. On the other hand, the droplet theory, which phenomenologically describes properties of spin glasses with short-range interactions, predicts that the SG order longer than a certain length scale L_h , called as the field-crossover length, is destroyed by h. Since L_h is finite for any finite h, it implies the absence of the SG phase under h.

We have been attacking this fundamental SG problem by means of numerical simulations, or *computational experiments*, on the Ising SG model. In our previous work on the field-shift aging protocol, we obtaind strong support for the presence of L_h and so the absence of the SG phase under h [1]. Recently we have carried out simulation on FCM processes by means of a standard Monte Carlo (MC) method [2]. The results are shown in Fig.1. The cooling rate is indicated by rn, namely, the system is cooled in temperature steps of 0.01*J*, while it is held at each temperature for a time, $t_{sty}=n$ MC steps. The FCM with slow cooling (large n) exhibits, with this enlarged scale of the ordinates, a peak at a certain temperature T^* . Numerically, we can measure the



Fig. 1. Field-cooled magnetization (FCM) and equilibrium magnetization (EQM) of the 3-dimensional Ising SG model with random nearest neighbor interactions which obey the Gaussian distribution with mean zero and variance J. Its SG transition temperature under h=0 in the thermodynamic limit is estimated as 0.95J. The field strength is represented by the corresponding Zeeman energy in unit of J.



Fig. 2. Halted FCM after slow and rapid cooling (sI-HFCM and rp-HFCM), and EQM at T_{hlt} below T^* . The QUFM (QUPM) is the time evolution of magnetization after instantaneous quench from completely polarized (random) states. In QUFM we can see its initial decrease up to a value smaller than EQM as well as its upturn to EQM.

equilibrium magnetization (EQM) by making use of methods, such as the replica MC method, by which we artificially accelerate the relaxation to equilibrium. Interestingly, FCM exhibiting a peak at T^* is smaller than EQM in a whole range of T simulated. Much more interesting phenomena are observed when the cooling process is halted at a temperature denoted as T_{hlt} . For $T_{\text{hlt}} > T^*$, the halted-FCM (HFCM) relaxes smoothly to EQM as expected. For $T_{\text{hlt}} < T^*$, on the other hand, HFCM after slow cooling changes in the opposite direction to EQM as shown in Fig. 2.

The peak structure in FCM and the peculiar relaxation process of HFCMs are observed also by real experiments on Ising spin glass Fe_{0.55}Mn_{0.45}TiO₃ [3] and other spin glasses, though EQM is not measured since the equilibration cannot be completed within the laboratory time window. A key idea to interpret these intriguing FCM phenomena is that, in states far from equilibrium, individual spins are thermally fluctuating under effective fields from their nearest-neighboring spins (plus h) without knowing their preferred direction in the destination, *i.e.*, in equilibrium. At T higher than T^* , SG short-range clusters grown in regions with less frustration are thermally blocked in a limited time scale of t_{sty} . As T is further decreased, clusters of smaller sizes become thermally blocked, and at T around T^* these clusters (of mean linear size L_{cl}) become in touch with others. Then spins on cluster boundaries feel new effective fields and they tend to align to the direction which increases the SG short-range order and so decreases FCM. This is our scenario [3] to interpret FCM decrease at $T < T^*$ and so a FCM peak at T^* . The FCM as well as HFCM continue to decrease so long as L_{cl} is significantly smaller than L_h . Our scenario ends up with speculation that, after the mean size of SG cluster reaches L_h , the equilibration of an assembly of SG clusters of mean size L_h starts to occur, making HFCM increase to EQM. Actually, such upturn of HFCM is observed in *real* experiments at T_{hlt} below but very close to T^* [3].

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Asymptotic Dimensional Reduction near a Quantum Critical Point

Kawashima Group

The principal effect of frustration is cancellation among competing couplings. In quasi-two-dimensional systems, when the frustration exists in inter-layer couplings, it tends to enhance independence of each layer. At finite temperature, however, this cancellation can never be perfect so as to make the system purely two-dimensional because thermal fluctuation generates effective net interlayer couplings. Even at zero temperature, the geometric frustration fails to decouple layers completely since the zero-point fluctuations restore the inter-layer coupling as discussed by Maltseva and Coleman. As a result, the critical phenomena in any real quasi-two-dimensional system belongs to the universality class of some three dimensional theory no matter how perfectly the frustration may seem to cancel the couplings. However, we find that this does not necessarily apply to the quantum critical point [1]. Namely, as one approaches to the quantum critical point, the fluctuation that introduces effective coupling dies out and pure two-dimensional behaviors dominate the system. The first experimental evidence of this phenomenon was found by measuring critical exponents of a field induced QCP in BaCuSi₂O₆ [2].

BaCuSi₂O₆ is a spin dimer system whose highly symmetric crystal structure gives it unique advantages for tackling the fundamental role of dimensionality in the field of quantum criticality. The material consists of layers of spin dimers stacked on top of each other. Each layer is a square lattice. However, the dimer plaquettes are staggered between consecutive layers, as shown in Fig. 1. This leads to geometrical frustration of the inter-layer antiferromagnetic interaction. That is, the antiparallel arrangement of spins within and between layers conflict. We consider quasi-two-dimensional anti-ferromagnetic Heisenberg model.

$$H = J_0 \sum_{i} \mathbf{S}_{i1} \cdot \mathbf{S}_{i2} + J' \sum_{(i,j),\alpha} \mathbf{S}_{i\alpha} \cdot \mathbf{S}_{j\alpha} + J_{\perp} \sum_{(i,j)} \mathbf{S}_{i1} \cdot \mathbf{S}_{j2}$$
$$(0 < J_{\perp} << J' << J_0)$$

Here, the first term represents the intra-dimer coupling whereas the second and the third terms the inter-dimer couplings. The second term is the coupling within a layer and the third the inter-layer coupling.

We study the quantum critical point by using the spinwave treatment and then examining magnon excitations [1].

• = an effective spin



Fig. 1. A schematic diagram representing the body-centered tetragonal BaCuSi₂O₆ crystal lattice. A dot represent a dimer formed from two Cu^{2+} S=1/2 spins. It is apparent that this lattice structure leads to geometrical frustration.



Fig. 2. [top] Phase boundary near the QCP measured in BaCuSi₂O₆ [2] compared to the curves obtained from a MC simulation [1]. [bottom left] Similar comparison of the magnetization for T = 30 mK and varying *H*, and [bottom right] for $H = H_c$ and varying *T*.

The effective coupling between layers turned out to be proportional to the number of excited magnons, which means that the fluctuation responsible for the effective inter-layer coupling dies out as we approach zero temperature and the quantum critical point can possess purely two dimensional characteristics. Interactions between either thermally excited or quantum condensed bosons induce a crossover to d = 3away from the QCP. Key to this result is the observation that zero-point phase fluctuations of the order parameter are suppressed near a chemical potential tuned BEC. The twodimensional value of the exponent characterizes the phase boundary near the zero temperature, in agreement with the experiment [2]. The agreement between the pure two-dimensional behavior and the real experiment is not only qualitative, but it is quantitatively good as can be seen in Fig. 2. The upper panel shows the critical field measured in the experiment on BaCuSi₂O₆ plotted with the phase boundary obtained by quantum Monte Carlo simulation assuming that the inter-layer coupling is zero (filled circles). Both the results show the two-dimensional quantum criticality that is characterized by the asymptotic linear dependence. The correspondence is further confirmed by other quantities: the field-dependence of the magnetization at zero (or very low) temperature (lower left panel), and the temperature-dependence of the magnetization at critical magnetic field (lower right panel).

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A New Record of Highest Magnetic Field, 630 T Generated by the Electro-Magnetic Flux Compression

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We have recently improved the primary coil of the electro-magnetic flux compression (EMFC) system, allowing it to generate a higher magnetic field with less energy injection and involving a more simplified coil preparation processes. Thus, we have renewed a record of generating highest magnetic field to 630 T.

The new coil system has a bent copper sheet inside a steel coil (Fig. 1). In the new coil system, the copper sheet bears the main primary current flow. The shape of the copper sheet is specially designed to improve the electric contact with the electrode of the current collector plate from the capacitor bank. The role of the steel coil differs from that in the old type ones. The steel coil in the new type is designed to provide sufficient mass to avoid deformation of the copper sheet during the liner acceleration.

Figure 1 shows the field pulses generated by EMFC, showing new world record achieving 630 T. The trace of the previous record is presented for a comparison, where the "feed-gap compensator" was adopted, and 622 T has been achieved as the indoor world record [1]. Figure 2 shows a high-speed photograph of the liner implosion. The cylindrical symmetry of the liner implosion was improved. The most important factor in generating high magnetic field by flux compression is the velocity of the liner. The velocity of the liner has also been improved to 2.4 km/s from the old



Fig. 1. The traces of magnetic field recently achieved by the EMFC. The background is a photograph of the new coil. 630 T is the new indoor world record achieved by the new coil.



Fig. 2. A high-speed photograph of the liner implosion process taken by a high-speed camera. (a) An implosion using the new coil, (b) An implosion using the old coil equipped with a feed-gap compensator.



Fig. 3. The image plot of the calculated temporal evolution of the current density and the temperature distribution in iron (a,b) and copper (c,d), respectively. The injected current J(t) used in the calculation is shown above each image plot.

record; 2.0 km/s. In the new coil system, the efficiency of the electric-magnetic energy transfer was estimated to be about 11 %, again much improved from the old one with about 5 %. We could have thus improved the efficiency of the electric-magnetic energy transfer more than two times.

It is considered that this arises from a difference in the current density distribution between the new coil and the old one. More specifically, it originates from the difference in the skin-depth. We have conducted the current density calculation from a combination of Maxwell equations and the heat conduction taking account of the Joule heating,

$$j = j_0 \exp\left(-\frac{x}{d} + i\left(\sqrt{\frac{\mu_0 \omega}{2\rho} x - \omega t}\right)\right), \quad (1)$$

with $d = \sqrt{\frac{2\rho}{\mu_0 \omega}}.$

Here, *d* denotes the skin depth which is a length representing a current density distribution.

$$c_{p}(T) D \frac{\partial T}{\partial t} = \lambda \frac{\partial^{2} T}{\partial x^{2}} + \rho(T) j^{2}.$$
 (2)

Here, c_p is the specific heat, D is the density, λ is the thermal conductivity and T is the temperature. We calculate the current density distribution and the temperature distribution in the case when the external current J(t) is injected to the metal plate. The results of calculation are shown in Fig. 3. Fig. 3(a) and (c) show the temporal evolution of the current density inside the primary coil made of iron and copper, respectively. The upper part of Fig. 3(a)(c) is an illustration of the current J(t) used in the calculation. Figures 3 (b) and (d) show the temporal evolution of the temperature distribution. It can be seen both from Fig. 3(a) and (c) that the current gradually penetrates deep into the bulk from the inner surface although the current is initially concentrated near the surface. As the time evolves, the peak of the current density also moves inside into the material. It can be seen in Fig. 3(b) or (d) that the temperature near the surface is much higher than the temperature inside the material. This causes the increase of resistivity near the surface and the decrease of the current density there. Thus, the peak of the current density gradually moves inside. Therefore, the diffusion of the current density in the iron coil is more prominent than that in copper one.

The liner is accelerated within the initial 30 μ s according to the pictures shown in Fig. 2. The current density distribution within this moment plays a crucial role on the liner motion and the symmetrical shape during the implosion. The effective feed-gap in the new coil is decreased to 6 mm at 30 μ s from 15 mm of the old iron one. It is considered that the difference of the current density distribution between iron and copper as shown above resulted in the difference of the shape of the liner during the implosion in Fig. 2. A rough insight into the current density distribution in our new primary coil has been obtained by our simple calculation.

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Metamagnetic Transition to *Poor* Conductor in BaVS₃

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In the hexagonal perovskite-type sulfide BaVS₃, consisting of one-dimensional (1D) spin chains with S = 1/2on V^{4+} along the *c* axis, several electronic instabilities and a series of symmetry-breaking have been found in spite of, at a first sight, simple structural and electronic characteristics at room temperature. This is due to the interplay among spin, orbital and charge degrees of freedom accompanied by spin frustration, orbital degeneracy, low dimensionality, strong covalency of sulfur, etc. The characteristic temperatures and the successive phase transitions are summarized as follows: (a) $T_{\rm S} = 240$ K, structural transition from hexagonal to orthorhombic, (b) $T_{\rm MI}$ = 70 K, metal-insulator (MI) transition with superlattice formation, (c) $T_{\rm X} = 30$ K, incommensurate magnetic ordering. In this report, we present high-field experiments of single crystalline BaVS₃ up to 70 T, which reveals a metamagnetic transition to a field-induced ferromagnetic state. A field-temperature phase diagram is proposed for BaVS₃ [1].

The high-field magnetizations up to 70 T have been measured by using non-destructive pulse magnets. Figure 1 shows magnetization curves at 4.2 K measured in the field parallel and perpendicular to the *c* axis. The magnetization increases linearly up to around $B_{\rm M} \sim 50$ T, and exhibits an



Fig. 1. High-field magnetization curves at 4.2 K in *B* parallel and perpendicular to the *c* axis. The low-field part at B < 20 T is magnified in the inset. A slight change in the curvature is seen around 10 T for both directions.



Fig. 2. *B-T* phase diagram of BaVS₃. AFI and PMI represent the antiferromagnetic insulator and the paramagnetic insulator, respectively. Broken and dashed-dotted lines indicate expected phase boundaries.

abrupt jump with a large hysteresis of about 8 T. At $B > B_M$, the magnetization shows a plateau with the magnitude of about 0.4 μ_B /f.u., which is approximately half of the saturation magnetization expected for V^{4+} (3d¹) with spin S = 1/2. A tiny anomaly, *i.e.*, the slight deviation from the linear relation, is observed around $B_X \sim 10$ T for both directions. Temperature dependences of field-induced (FI) anomalies are summarized as the B-T phase diagram shown in Fig. 2. Being different from the appreciable temperature dependence of the high-field transition point, the low-field anomaly shows a weak temperature dependence and suddenly disappears at T_X . We believe that the metamagnetic transition at $B_{\rm M}$ is not of classical spin flip but corresponds to an essential change in the electronic state, because (1) the anisotropy in the transition field is weak, (2) the transition is accompanied by a large hysteresis, suggesting strong spin-lattice coupling, and (3) the transition is observed even in the spin disordered state at $T_X < T < T_{MI}$. Although one may expect that the FI transition at $B_{\rm M}$ corresponds to the temperature-induced MI transition, the nature in the field above $B_{\rm M}$ is much different form that at $T > T_{MI}$. The moment appeared above B_M is nearly half of S = 1/2, which is comparable to the ordered moment 0.5 $\mu_B/f.u.$ estimated from neutron experiments at zero field [2]. This means that, in contrast to the spin reduction at $T_{\rm MI}$, the spin retains the magnitude through both the transitions at B_X and B_M .

Recently, Kézsmárki et al. [3] proposed that 1D metallic (good) transport of A_{1g} electrons is superimposed on the isotropic hopping (bad) conduction of localized E_g electrons. One of the possible explanations of the metamagnetic transition at $B_{\rm M}$ is that the gapped $A_{1\rm g}$ state is not affected appreciably by the field of 50 T, while the E_g orbitals are reconstructed to a quasi-degenerate state by the application of the field. To see the nature of the $B_{\rm M}$ - $T_{\rm MI}$ boundary, we investigated a field dependence of the conductivity and its anisotropy at 35 and 60 K. The experiment reveals that only the resistivity in the c plane decreases at $B_{\rm M}$, however, the reduction is not huge like a MI transition, suggesting that the FI transition is not a pure MI transition. It is, therefore, reasonable to conclude that the FI state is a poor conductor only in the E_g bands. This interpretation also intimates the presence of another phase boundary to a real metallic state at

higher fields (broken line in Fig.2). The inside of the B_X-T_X boundary should be a long-range-ordered antiferromagnetic insulating (AFI) phase. It should be noted, however, that the anomaly at B_X is faint, and that the entropy is little consumed at T_X . These results indicate that the strong antiferromagnetic correlation persists even in a paramagnetic insulating (PMI) state outside the B_X-T_X boundary. On the other hand, it is hardly acceptable that the external field induces the dynamic PMI state from the AFI state. The width of dM/dB peak at B_M is, in fact, broadened markedly above T_X , suggesting the presence of another unidentified phase boundary or crossover, which discriminates static and dynamic states (dashed-dotted line in Fig. 2).

In conclusion, we have established the *B*-*T* phase diagram of BaVS₃. Being different from the metallic state above $T_{\rm MI}$, the FI state above $B_{\rm M}$ is probably a poor conductor, where the conductivity is recovered only in the $E_{\rm g}$ bands and the gapped $A_{\rm 1g}$ state remains open. The detection of the new phase boundary to the real metallic state remains for further studies.

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