The Institute for Solid State Physics The University of Tokyo



Activity Report 2007







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ISSP

Activity Report 2007

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ISSP Activity Report 2007

Preface

This booklet constitutes the annual report of the activity of the Institute for Solid State Physics (ISSP) of the University of Tokyo for the fiscal year 2007. The ISSP was established in 1957 as a joint-use central research institution in the field of basic materials science, so that the year 2007 was the 50th anniversary.

In the late 90's, ISSP made a big decision to leave the Roppongi campus after its 40 years of operation there in order to seek a fresh restart in the new Kashiwa campus. In preparation of the restructuring and relocation, the ISSP underwent its first major external evaluation by an international review committee in 1995. The comments and suggestions given by the review committee were highly valuable in envisioning the future of the ISSP in Kashiwa. The construction of research building started in 1997 and the first group of laboratories began research operation in Kashiwa in April 1999. The relocation of the main body of the ISSP followed and the full operation in Kashiwa started in April 2000.



In the fall of 2005, the ISSP had the second major external evaluation by an international review committee. In the review, evaluation was given on the research accomplishments, especially those utilizing new facilities installed at the time of relocation. I believe it is fair to say that the scientific achievements during the first 5 years of research activity in Kashiwa were highly praised by the reviewers. Valuable comments and suggestions were also given on the future research directions and the betterment of the ISSP management.

In November 2007, ISSP held the 50th anniversary and a commemorative symposium in the old Roppongi campus, which has now been renovated as the National Art Center and the National Graduate Institute for Policy Studies. For many old-timers, this was a kind of "home-coming" event. In the symposium, the "past, present and future" of ISSP and materials science were discussed.

In addition to the various on-going research activities, there are a few near-future projects ranging from the actual construction stage to the planning stage. The one currently in the final stage of construction is the long-pulse high magnetic field laboratory, which, when completed, will offer a versatile platform for experiments in high magnetic field, and will constitute a comprehensive International MegaGauss Science Laboratory for high magnetic field research together with the existing short-pulse megagauss field installation. Another project vigorously promoted is the Advanced Coherent Light Laboratory which is envisaged as a laser facility enabling experiments in the unexplored realm of short wavelength, ultrafast and high peak power.

The ISSP is now taking a step forward to its second 50 years. It is a good occasion for us to reflect on the mission, past accomplishments, and future strategy of the ISSP, so as to keep playing the leading role for the advancement of basic science in the relevant fields by the synergy of the in-house staffs and the research communities.

> June, 2008 Yasuhiro Iye Director Institute for Solid State Physics The University of Tokyo

Research Highlights

A Novel Ordered Phase in the Frustrated 2D Spin System SrCu₂(BO₃)₂ under High Pressure

Takigawa, Y. Ueda, and Uwatoko Groups

A variety of exotic quantum phenomena has been discovered in SrCu₂(BO₃)₂, a quasi two-dimensional frustrated spin system with a dimer-singlet ground state (Fig. 1ab), most notably, the sequential magnetization plateaux at 1/8, 1/4, and 1/3 of the saturation. These plateaux accompany superlattice of triplets that breaks the translational symmetry of the crystal, as observed by NMR experiments [1].

Compared with the behavior in high magnetic fields, little has been known about the properties under high pressure, although pressure would be a powerful tool to investigate quantum phase transitions expected for spin systems with such a geometry. We have performed nuclear magnetic resonance (NMR) experiments on ¹¹B nuclei in a single crystal of SrCu₂(BO₃)₂ under pressure up to 2.4 GPa, which revealed a magnetic phase transition resulting in a novel type of valence-bond-solid (VBS) order. A pistoncylinder type pressure cell was mounted on a doubleaxis goniometer, which enabled precise alignment of the magnetic field along arbitrary crystalline directions.

The NMR spectra at 2.4 GPa in the field of 7 T along the *c*-axis are shown in Fig. 2 at various temperatures (*T*). A sharp single line is observed at high *T* consistent with the crystal symmetry. The line, however, splits gradually below 30 K. The angle dependence of the spectrum indicates that the splitting is due to loss of C_4 symmetry of the crystal, which makes the two orthogonal dimer sublattices inequivalent (solid and dashed blue lines in Fig. 1a). With decreasing temperature, each line further splits into one sharp and one broad lines below 3.6 K. The magnetic hyperfine shift (*K*) of the sharp line shows an activated *T*-dependence indicating an excitation gap, while the broad line shows gapless behavior with nearly constant *K* (Fig. 3a). The clear



Fig. 1. The magnetic layer of $SrCu_2(BO_3)_2$ viewed along (a) the *c*-direction and (b) the [110]-direction. The Cu^{2+} ions with spin 1/2 form orthogonal dimer sublattices as shown by the solid and the dashed blue lines. (c) Proposed VBS order of alternating magnetic (shaded) and non-magnetic (non-shaded) dimers.



Fig. 2. NMR spectra for H // c at various temperatures. A gradual line splitting below 30 K is followed by another sudden splitting of each line into one sharp and one broad lines below 3.6 K.



Fig. 3. (a) Temperature dependence of the magnetic hyperfine shift. The black open squares show the data for unsplit lines at high T, while the blue and red open squares represent the shifts for split lines in the intermediate T-range. The filled symbols indicate the shift in the ordered phase. For comparison, the data at ambient pressure are shown by the green dots. (b) Field-temperature phase diagram obtained from the NMR data.

singularity in both K(T) (Fig. 3a) and the susceptibility indicates a well defined magnetic phase transition.

The detailed investigation of the angle dependence of the spectra allowed us to conclude that each dimer sublattice develops a superstructure with alternation of magnetic and non-magnetic dimers, resulting in a doubled unit cell (Fig. 1c). The magnetic dimers appear to restore a finite gap at low magnetic fields below 2 T. Hence the ground state at zero field would be a novel type of valence-bond-solid state with staggered dimer correlation, which breaks the translational symmetry but does not show any spontaneous

magnetization. The transition temperature decreases with magnetic field (Fig. 3b), suggesting a quantum phase transition near 18 T. Further experiments to clarify the nature of this totally unexpected ordered phase are in progress.

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Gap Determination of the Heavy Fermion Superconductor URu₂Si₂ by Specific Heat Measurements in Magnetic Fields

Sakakibara Group

The tetragonal compound URu₂Si₂ has attracted much interest, primarily because it exhibits an exotic ordering at $T_0=17.5$ K whose order parameter is still an issue of intense debate and referred to as a "hidden order parameter" [1]. This hidden order coexists with superconductivity that sets in below $T_c=1.4$ K [2]. Regarding the superconducting properties of this system, the existence of nodes in the superconducting gap has been reported in several experiments including the specific heat, NMR and NQR measurements. The pairing symmetry, however, has remained unidentified. In order to examine the gap structure of the superconducting state, we have carried out field-orientation dependent specific heat C(H) measurements on a single crystal of URu₂Si₂ ($T_c = 1.3$ K, 100 mg weight) [3].

Figure 1(a) shows C(H) of URu₂Si₂ for various orientations between the c- and a-axes. It is remarkable that the anisotropy of C(H) is weak at low fields, but becomes pronounced when H exceeds about 1 T where the effect of anisotropy in the upper critical field H_{c2} becomes significant. Accordingly, the field dependent part of the specific heat C_H as a function of H/H_{c2} becomes strongly field-



Fig. 1. (a) Field variations of the specific heat of URu₂Si₂ obtained at 0.34 K for various field-orientations between c and a axes. (b) Fielddependent part of the specific heat for the two principal directions plotted as a function of H/H_{c2} .



Fig. 2. Schematic gap structures for the superconducting state of URu₂Si₂. (a) is the gap structure suggested by the C_H measurements. Two point nodes are located in the z direction. Group theoretical considerations require an additional line node as shown in (b), which is probably invisible due to the light effective mass of the quasiparticles.

orientation dependent at low fields as shown in Fig. 1 (b). When $H \parallel a$, C_H exhibits a steep increase at low field with a clear downward curvature, similar to the $H^{1/2}$ behavior of nodal superconductors due to the "Doppler effect" of moving quasiparticles (QPs) near the gap nodes [4]. As for $H \parallel c$, however, C_H almost linearly increases at low fields, which is the behavior expected for a full-gap superconductor. More generally, this means that there is no nodal QP that is Doppler-effect active for this field orientation. These two qualitatively different field dependences of C_H can be reconciled with each other if point nodes are located along the z directions as depicted in Fig. 2 (a). In such a case, the nodal QPs cannot contribute to the zero-energy density of states when $H \parallel c$, because the momentum of those QPs is perpendicular to the superfluid velocity (circulating supercurrent around the vortices), making the Doppler effect inactive.

As can be seen in Fig. 1 (b), C_H for $H \parallel c$ exhibits a distinct upturn above $H \sim 0.4H_{c2}$. In fact, a linear extrapolation of the low field data of C_H (the dotted line in the figure) reaches the normal state value at $H/H_{c2} \sim 1.2$. This behavior is in contradiction with the case of ordinary (nodal as well as full-gap) superconductors where the similar extrapolation reaches the normal-state value at $H/H_{c2} < 1$. This unusual behavior of C_H in URu₂Si₂ can be explained by a strong Pauli paramagnetic depairing effect that suppresses H_{c2} to the value well below the orbital limiting field [3].

These observations lead us to conclude that the pairing symmetry of the superconducting state in URu₂Si₂ is even parity (singlet pairing) with point nodes in the z (polar) direction (Fig. 2 (a)). Group theoretical considerations for a tetragonal crystal tell us that a possible even parity gap function with linear point nodes is $k_z(k_x+ik_y)$. This gap function, however, possesses a line node on the equator $(k_z=0)$ (Fig. 2 (b)), whose presence was not confirmed in our experiment. We believe that this discrepancy comes from Fermi surface topology of the present system. It should be noticed that C(H) measurements mainly probe the quasiparticles in the heaviest band. If the line node in question is located on a band with a light effective mass, it would not be detected by the C(H) measurements. While we have to consider realistic Fermi surfaces for conclusive evaluation of the gap structure, linear point nodes in the polar direction are quite plausible in URu₂Si₂.

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Electroluminescence Quenching Caused by a Spin-crossover Transition

Tajima Group

The spin-crossover transition in metal-coordinate system interests many researchers from the view pint of various functionalities such as color change and photo response (LIEST) [1]. In order to utilize them in an electric device, thin-film fabrication is necessary. In this respect, we have fabricated a thin film of $[Fe(dpp)_2](BF_4)_2$ (dpp = 2,6-di(pyrazol-1-yl)pyridine) by employing the spin-coating technique. This compound exhibits a transition around 260 K in a single crystal [2]. The fabricated films were ~30 nm in thickness with roughness less than 5 nm. Temperature dependence of the magnetization of the films exhibits a spin-crossover transition around 260 K. We also observed changes associated with the spin-crossover transition in optical absorption spectra [3].

Very recently we fabricated organic light-emitting diode using [Fe(dpp)₂](BF₄)₂ as the host compound and chlorophyll a as the guest. We found an electroluminescence quenching caused by the spin-crossover transition. The phenomenon is reproducible for thermal cycles. *This is the first observation of a switching phenomenon in an organic thin film device accompanying the spin-crossover transition* [4].

Figure 1 shows the device architecture and the structures of molecules used for device fabrication. The thickness of the emissive layer was approximately 50 nm. The



Fig. 1. The device architecture, molecular structures of $[Fe(dpp)_2]^{2+}$ and chlorophyll a, and the image of thin film of $[Fe(dpp)_2](BF_4)_2$.



Fig. 2. Electroluminescence spectra of the ITO/chlorophyll $a:[Fe(dpp)_2](BF_4)_2/Al$ device at different temperatures under the applied voltage of 3.5 V. The inset shows the temperature dependence of the EL intensity at 689 nm.

roughness of the surface was less than 5 nm.

Figure 2 shows the EL spectra at different temperatures under the applied voltage of 3.5 V, and the inset depicts the temperature dependence of the EL intensity at 689 nm where the emission maximum was observed. The EL emission from the fabricated ITO/chlorophyll a: [Fe(dpp)₂](BF₄)₂/Al device under the applied voltage of 3 V was clearly observed at 300 K, and the spectrum was the same as that of the single-layer device with chlorophyll a [5]. However, as the temperature decreased to 200 K, the EL spectrum of chlorophyll a could not be observed; heating to 300 K enabled the spectrum to be obtained again. This ON-OFF switching of the EL emission is reproducible for the heating and cooling processes. As the temperature decreases, the intensity of the EL emission decreases, and around 260 K, the intensity becomes comparable with that of the background level (< 260 K).

Since the temperature of 260 K is almost the same as the spin transition temperature of [Fe(dpp)₂](BF₄)₂ complex, it is considered that the EL quenching at 260 K is relevant to the spin transition. One possible origin is an energy transfer from excited chlorophyll a to $[Fe(dpp)_2](BF_4)_2$. As a result of the spin transition, the energy level of [Fe(dpp)₂](BF₄)₂ should change. If the energy level shifts from above to below that of the excited chlorophyll a, the energy transfer that leads to the nonradiative decay process can occur. In order to examine this possibility, we measured the photoluminescence (PL) spectra for the chlorophyll a: [Fe(dpp)₂](BF₄)₂ film at 300 and 200 K. Although the PL intensity observed at 200 K was weaker than that at 300 K, the PL spectrum of chlorophyll a could be obtained at 200 K in contrast to the EL spectra vanishing. This result rules out the possibility that the quenching of the EL emission from chlorophyll a is caused by the energy transfer, and suggests the disappearance of the excited chlorophyll a in the EL device. We speculate that carrier injection efficiency to chlorophyll a is drastically changed as the spin transition of [Fe(dpp)₂](BF₄)₂ occurs. In other words, in the low-temperature region where [Fe(dpp)₂](BF₄)₂ is at a low-spin state, injected carriers pass through the insulating layer via [Fe(dpp)₂](BF₄)₂ without generating the excited state of chlorophyll a. Detailed studies are under way.

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Giant Nonlinear Conduction and Electric-Field-Induced Metastable State in an Organic Conductor β-(meso-DMBEDT-TTF)₂PF₆

Mori Group

In the past few decades, nonlinear conductions in organic conductors have been extensively studied in neutral-ionic transition complexes, Spin-Peierls materials, Mott insulators, π -d systems, and others. Recently, as a new type of nonlinear conductors, the charge ordered (CO) organic materials are attracting much attention because they require low voltage to activate their nonlinear conductions. The low activation voltage enables us to effectively exclude the problems of Joule heating and may help studies on intrinsic nonlinear conduction. Moreover, the peculiar thyristor-like I-V characteristics and DC-AC converter effect of θ -(BEDT-TTF)₂CsM'(SCN)₄ (M'=Zn, Co) once again promoted the study of nonlinear conduction phenomena, especially in CO materials [1]. To investigate the relationship between charge ordering and dielectric response and to study nonlinear conduction in CO materials, we carried out measurements on dielectric constant and I-V characteristics in an organic CO material, β -(meso-DMBEDT-TTF)₂PF₆ [2-5].

Figure 1 shows *I-V* curves obtained by the *I*-driven 4-probe method. Nonlinear conduction is observed in a wide temperature range below 100 K. Especially, the negative differential resistance (NDR: dV/dI < 0) can be seen below about 70 K. It should be noted that the voltage drop pauses around 20 A/cm² and then the small NDR appears around 40 A/cm². To clarify the singularity in its *I-V* curves, we fit the results of the 4-probe measurement by the phenomenological equation:



Fig. 1. *I-V* curves obtained by the *I*-driven 4-probe method for β -(*meso*-DMBEDT-TTF)₂PF₆. Solid lines are the fit to eq. (2).



Fig. 2. Apparent sample resistivity E_{sample}/J as a function of V_{circuit} for β -(*meso*-DMBEDT-TTF)₂PF₆.



Fig. 3. Time-resolved sample voltage V_{sample} at 60.8 K for β -(meso-DMBEDT-TTF)₂PF₆.

The first term reflects the ordinary Arrhenius-type temperature dependence of conductivity. The second term means the current-induced enhancement of conductivity. By introducing the threshold current density J_T [$dE/dJ(J=J_T)=0$] we can rewrite this equation as follows:

$$\sigma(J,T) = \sigma_0[1 + (J/J_T)^n/(n-1)]$$
(2)

where $\sigma_0 = \sigma_1 \exp(-\Delta/T)$. We fitted the experimental data with the fitting parameters σ_0 , $J_{\rm T}$, and n in the range of $J \leq 10$ A/cm². From the fitting, we obtained $J_{\rm T} = 2.5$ -4.7 A/cm² and n = 1.8-2.0. This n value is larger than ordinary nonlinear conductors with the n value of ~1.5. The solid lines in Fig. 1 show the fit to eq. (2). We can see from the figure that the experimental data deviate upward from the fitting function in the high-current region. According to eq. (2), the conductivity increases with the power of J, which expects the linear behavior with the slope of n in the larger J region. However, the slope varies with the J value in the range of $J \geq 10$ A/cm² and cannot be determined uniquely.

To compare with the results of the V-driven 2-probe method, we plotted the apparent sample resistivity E_{sample}/J obtained from the 2-probe method as a function of V_{circuit} as shown in Fig. 2. At 55.5 K, for example, the resistivity shows a steep drop by a factor of 10^3 at the threshold voltage of 4 V. Such a giant nonlinear conduction with a low threshold voltage is similar to that of θ -(BEDT-TTF)₂CsCo(SCN)₄ and could be attributed to the melting of the CCO (checkerboard CO). It is also noted that the subsequent melting process occurs even after the giant resistivity drop above 4V.

To further investigate the peculiar two-stepped I-Vcharacteristics, we monitored the sample voltage under the pulsed circuit voltage using an oscilloscope (Tektronix TDS2012B). The results are shown in Fig. 3. If the circuit voltage V_{circuit} is small, the sample voltage V_{sample} keeps a constant value near V_{circuit} owing to its large resistivity $(R\sim1\times10^5$ ohm, $\rho\sim80$ ohm cm). When V_{circuit} reaches about 2 V, V_{sample} begins to drop suddenly with some delay after the pulse application and the larger V_{circuit} causes the faster drop of V_{sample} . In the short time span just after the steep voltage drop, we can see a terrace of low voltage (t = 1ms, $V_{\text{sample}} = 0.9 \text{ V}$ at $V_{\text{circuit}} = 3.00 \text{ V}$, for example) before dropping into the more conductive state. Such a non-monotonus response has never been observed in other materials studied so far, and this plateau would be the reason for its peculiar two-stepped NDR. Another noticeable point is that the LR-CCO (longrange-CCO below 70 K) shows a two-stepped drop with the metastable plateau, while the SR-CCO (shortrange-CCO above 70 K) gives a small, monotonous drop.

In conclusion, we observed the formation of LR-CCO as a steep increase in the frequency-dependent dielectric constant at 70 K. In the transport measurement, (1) the nonlinear *I-V* curve below 100 K, the peculiar negative resistance around 70 K, and a giant resistance drop by a factor of up to 10^3 under 4 V at 55.5 K were observed, and in the time-resolved measurement technique (2) a two-stepped sample voltage drop with a transient plateau occurred below 70 K. This indicates that the melting of SR-CCO gives a small and monotonous voltage drop, whereas the LR-CCO changes drastically to a temporary plateau before dropping to the most conductive state. Especially, this plateau below 70 K implies a new metastable state that can be induced only by the electric field.

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High Temperature Superconductor Micro SQUID Magnetometer for Magnetization Measurement of a Micro Scale Magnet

Mori and Ishimoto Groups

So far there are several reports on the HTS SQUID techniques, where the bare SQUID loop as well as the micro SQUID system is used as the detection coil. The scanning SQUID microscope (SSM) can observe stray magnetic field images produced by small wires of ~20 μ m in diameter. However, these measurements were made only in fixed



Fig. 1. The photoimage of the fabricated HTS SQUID.

fields less than 5-mT, not suitable for the magnetization reversal measurements. Here, we report on the operation of HTS micro SQUID with a thinner film and a smaller loop than SSMs to enable measurements while sweeping the magnetic field produced by a 3D magnetic field coil system. The hysteresis loop of micro ferrimagnetic crystal has successfully been measured in any direction of magnetic field up to 0.12 T between 30 and 70 K [1, 2].

A dc micro SQUID with grain boundary Josephson junctions (GBJJs) was fabricated according to the standard procedure. By using a pulsed laser ablation technique, epitaxial 92 nm thick YBa2Cu3O7-& (YBCO) films were deposited onto a [001]-oriented SrTiO3 (STO) bi-crystal substrate $(10 \times 10^2 \text{ mm}^2)$ with misorientation angle of 24°. The YBCO film deposition was followed by in-situ sputter deposition of 70 nm-thick Au layer for electrical contact and protection during the subsequent processes. The dc micro SQUID was formed by using the photolithography and the ion milling dry etching techniques to have a hole of $4 \times 9 \ \mu m^2$ and two Josephson junctions with 2 μm in width (W) across the bi-crystal line (Fig. 1). The fabricated chip was mounted on a copper holder with a thermometer (Lakeshore CERNOX 1030) and a heater to control the temperature from 4.2 K to 300 K. The holder is supported from the top of vacuum can (VC) by several rods of phenolic resin (PPS) for moderate cooling and efficient temperature



Fig. 2. Schematic diagram of the measurement probe unit with a 3D magnetic field coil system.

control. The 3D magnetic coil system (B_1 , B_2 , and B_3) placed outside the VC consists of three mutually orthogonal coils. This allows accurate sweeping of the magnetic field in the desired orientation (Fig. 2).

The present system was used to measure the magnetic hysteresis of a micron-sized magnetic material. In the pointdipole approximation, a dipole moment μ (JT⁻¹) produces a flux at the SQUID loop:

$\phi = B_{\rm s} A_{\rm eff} = 3\mu \,\mu_0 \,A_{\rm eff} \,\sin\theta \cos\theta \,/\, 4\pi |\mathbf{r}|^3$

Here, B_s is the magnetic flux density from the sample placed at the center of the SQUID loop, A_{eff} =77.4 μ m², μ_0 the free-space permeability constant, r the vector joining the sample to the SQUID loop, and θ the angle between r and the direction of the dipole moment. The sample here is a molecular ferrimagnet: [Mn₂(H₂O)₂ (CH₃COO)][W(CN)₈]·2H₂O with $T_c = 40$ K. Panels (a) and (b) in Fig. 3 represent I_c vs B_2 at 35 and 50 K, respectively. At 35 K, the SQUID modulation period precipitously drops as the field approaches 0 T, while at 50 K it is almost constant



Fig. 3. I_c vs B_2 plots at 35 K (a) and 50 K (b) with the sample mounted on SQUID II. The black and gray lines show the field-reducing and -increasing processes, respectively. The inset to (a) gives details around 0 T. The black-circle and gray-triangle markers in (c) show the magnetization curves of the sample deduced by counting the number of the detected flux depending on the applied magnetic field at 35 and 50 K, respectively. At each temperature the filled (open) markers denote the curves in the field-reducing (-increasing) processes. The behavior in the range of -24 and -10 mT in the field-reducing (d) and -increasing processes (e), respectively.

over the whole field range. Furthermore, as is seen in the inset of Fig. 3(a), the central peaks in the Fraunhofer like pattern at 35 K exhibit negative- and positive-field shifts in the field-decreasing and -increasing process, respectively. There is no such behavior at 50 K. This may imply a generation of remanent magnetization at 35 K. Second harmonic components appear in the modulation in the range of -24 to -10 mT in Fig. 3(d) and (e). The period is likely reduced by a factor of two compared with a normal one. It leads to $1/2\phi_0$ period wherever the second harmonic component appears. In this way the magnetization curves are obtained within an accuracy of less than $1/2\phi_0$. There can be seen a clear contrast between the curves above and below T_c in Fig. 3(c). The magnetization curve at 50 K is linear with respect to the applied magnetic field. It is mainly from the stray field since the paramagnetic component in the sample is expected to be negligible. Thus, we can extract the sample magnetization curve at 35 K (the saturated value of ~40 ϕ_0 around 30 mT) by subtracting the 50 K curve from 35 K one.

In summary, we have developed an HTS micro SQUID magnetometer combined with a 3D magnetic field coil system that is designed for investigating the magnetization reversal of tiny magnetic materials. It can be operated in the temperature range between 30 and 70 K (an improvement over LTS micro SQUIDs) and under the external field up to at least 0.12 T. The anisotropic behavior of hysteresis loop has successfully been measured for a micro crystalline molecular ferrimagnet. This is the first time that a HTS micro SQUID under magnetic field up to above 0.1 T. The present sensitivity is an order of magnitude higher than MPMS but far below the submicron Hall magnetometer operable in similar temperature range. If the distance between the sample and the SQUID loop were reduced by a factor of ten in the present case, the sensitivity would be expected to increase roughly thousand times as much as now.

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2D Frustrated Magnetism vs. 3D Spin Order in the Chalcogenide Triangular Antiferromagnets NiGa₂S₄, FeGa₂S₄, and Fe₂Ga₂S₅

Nakatsuji Group

Geometrically frustrated magnets have attracted great interest for the possible exotic ground states and phase transition phenomena. The two dimensional triangular lattice is one of the simplest forms of a geometrically frustrated lattice with a single magnetic ion in a unit cell.

NiGa₂S₄ is the first example of S = 1 two dimensional (2D) antiferromagnets on an exact triangular lattice [1].



Fig. 1. (a) Four sublattices of the buckled honeycomb lattice formed by the Fe-S-Fe straight bonds in the bilayer of Fe₂Ga₂S₅. (b), Scaling in the T^2 dependence in the magnetic part of the specific heat C_M for NiGa₂S₄ (S = 1; $\Theta_W = 80$ K) and FeGa₂S₄ (S = 2; $\Theta_W = 160$ K) at 0 T in full logarithmic scale (c) *ab*-plane and *c*-axis components of the susceptibility χ for FeGa₂S₄ and Fe₂Ga₂S₅ under B = 0.1 T. Inset $\chi(T)$ and $d\chi/dT$ of Fe₂Ga₂S₅. The vertical lines indicate the Néel point T_N .

Despite an antiferromagnetic coupling with the Weiss temperature $\Theta_W = 80$ K, no antiferromagnetic or canonical spin glass ordering has been observed down to 0.08 K [1, 2]. Instead, recent nuclear-quadrupole-resonance and muon-spin relaxation measurements have revealed unconventional freezing phenomena that set in below $T_f \sim 10$ K and have a wide temperature critical fluctuation regime down to 2 K [2, 3]. Very interestingly, below $T_f \sim 10$ K, it develops a short-range noncollinear order with 2D gapless linearly dispersive excitations characterized by a T^2 dependence of the specific heat [1]. In order to explain the origin and stability of the 2D coherent behavior in the frozen spin-disordered state, various theoretical proposals have been made. Experimentally, it is highly important to clarify the effect of spin size change, bilayering, and geometrical frustration, and to see if any spin order can be realized in the related compounds.

Thus, we have made the single crystal study of the isostructural single-layer triangular antiferromagnets, NiGa₂S₄ and FeGa₂S₄, and the homologous bilayer triangular antiferromagnet Fe₂Ga₂S₅ [4]. We have succeeded in growing single crystals of NiGa₂S₄ for the first time and found properties fully consistent with those for polycrystalline samples. Interestingly, the magnetic properties of FeGa₂S₄ bear strong resemblances to those of NiGa₂S₄ despite the fact that Fe^{2+} has twice larger S = 2 spin than S = 1 for Ni²⁺. Both compounds have basically Heisenberg spins, and form frozen disordered state (see Figure 1). In addition, the specific heat is insensitive to the field and shows a T^2 dependence, which scales to that of NiGa₂S₄ (see Figure 1(b)). The similarities strongly suggest that the 2D coherent behavior in the frozen spin-disordered state has the same underlying mechanism. In contrast, a clear antiferromagnetic transition is observed for the bilayer system Fe₂Ga₂S₅, whose dominant antiferromagnetic bonds most likely form an unfrustrated honeycomb lattice (see Figure 1(a) and (c)). These results suggest that the geometrical frustration of the single-layer triangular lattices stabilizes the spin-disordered state, probably associated with 2D antiferromagnetic ordering [4].

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Nonperturbative Approaches to Correlated Transport through Quantum Dots under a Bias Voltage

K. Ueda Group

One of challenging problems in mesoscopic physics is to understand many-body effects in a nonequilibrium situation. A central issue is the Kondo effect at quantum dots. By applying a finite bias voltage, quantum dots enable us to investigate the nonequilibrium Kondo effect. We have employed the perturbative Keldysh Green function method, by which it has been settled whether the Kondo peak splits or not [1]. Toward full understanding of electron correlation effects at a steady state, nonperturbative treatments are also required. In recent years we have addressed this issue from two aspects.

A. Shot noise and nonequilibrium Kubo formula based on Keldysh formalism for mesoscopic systems

As for the Keldysh formalism applied for mesoscopic systems, we have examined an adiabatic switching-on of the perturbation term. We have proven that the procedure corresponds to taking the invariant part introduced by Zubarev. As a consequence the density matrix becomes one of MacLennan-Zubarev forms. This concept enables us to derive a general form of differential conductance, given by the sum of the current-current correlation function and the non-trivial current-charge correlation function. This general-



Fig. 1. The differential conductance under a magnetic field for U/t=2, where t is the hopping amplitude in the leads, U the Coulomb energy at the dot, V the bias voltage and \tilde{h} the Zeeman energy. It is well known that the split peak is located at the Zeeman splitting (twice of the Zeeman energy), and our result shows an excellent agreement.



Fig. 2. The expectation values of the double occupation operator taken by the steady state wave function for U/t=2. Here we can see a non-monotonic behavior for relatively small magnetic field.

ization may be called as the nonequilibrium Kubo formula.

It is well known that the current-current correlation function gives noise power at zero frequency. The generalized Kubo formula leads us to define shot noise by the non-trivial current-charge correlation function. This fact is confirmed in case of the noninteracting Anderson model. The nonequilibrium Kubo formula have thus provided a new perspective on the shot-noise study [2]. With the resulting formula of shot noise, we are currently studying the shot noise in the Kondo effect at quantum dots.

B. Time-dependent DMRG study on quantum dot under a finite bias voltage

Apart from Keldysh formalism, it seems that there are only few numerical studies on quantum dot under a finite bias voltage, due to a lack of reliable numerical techniques for nonequilibrium problems. One possible numerical approach is the adaptive time-dependent density matrix renormalization group (TdDMRG) method. In this method, the ground state wave function is calculated with the usual DMRG method initially. Then the time evolution of the wave function due to the slowly changing bias voltage between the two leads can be calculated by using the TdDMRG technique. Even though the system size is finite, the expectation value of the current operator shows steady like behavior for a finite time interval, in which the system is expected to resemble the real nonequilibrium steady state of the infinitely long system.

Taking this approach, we have shown that thus calculated wave function in the time interval contains accurate information on the properties of real steady state of the infinite system in a wide range of bias voltage [3]. We have successfully reproduced the splitting of the zero bias peak in the differential conductance, Fig. 1. Moreover, an anomalous behavior has been observed in the expectation values of the double occupation operator at the dot as a function of bias voltage, Fig. 2. This method is applicable also to other nonequilibrium problems of mesoscopic systems.

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Importance of Nonlocality in the Dynamical Exchange-Correlation Kernel in the Time-Dependent Density Functional Theory

Takada Group

The time-dependent density functional theory (TDDFT) is a powerful tool for studying excitations in atomic, molecular, and condensed-matter systems. In the linear-response regime, the key quantity of TDDFT is the dynamical exchange and correlation (xc) kernel $f_{xc}(\mathbf{r},\mathbf{r}';\omega)$, defined as the Fourier transform of $f_{xc}(\mathbf{r},\mathbf{r}';t-t')$ which is given by the functional derivative of the Kohn-Sham xc potential $V_{xc}(\mathbf{r},t)$ with respect to the electron density $n(\mathbf{r}',t')$. In actual implementation of the theory, we need to resort to some approximation to this quantity. In the absence of its detailed information, the adiabatic local density approximation (ALDA) has been adopted in virtually all calculations so far.

Some years ago, we derived a rigorous formula for obtaining the experimentally observed stopping power, -dE/dx, of solids for slow ions in the framework of TDDFT [1], but it turns out that the formula provides a nonzero value for the friction coefficient (or -dE/dx divided by v the velocity of incident ions in the limit of $v \rightarrow 0$) in the homogeneous electron gas with the limit of zero electron density (or in the vacuum), if we adopt ALDA for $f_{xc}(\mathbf{r},\mathbf{r}^*;\omega)$. This contradictory result urges us to develop a better approximation scheme for this quantity beyond the level of ALDA.

In pursuit of such a scheme, we have reformulated the problem of the stopping power in the framework of the time-dependent current density functional theory (TDCDFT) [2] to find a new interesting rigorous relation between $f_{xc}(\mathbf{r},\mathbf{r}';\omega)$ and the tensorial xc kernel, the key quantity



Fig. 1. Comparison between experiment on the friction coefficient of aluminum for slow ions (characterized by the atomic number) obtained at the incident velocity v = 0.5 atomic units (the solid squares) and theory on the same quantity of the homogeneous electron gas with the electron density equivalent to the valence electron density in aluminum in the limit of zero ion velocity ($v \rightarrow 0$). The open circles, the open triangles, and the open squares correspond, respectively, to the results in our new approximation scheme (LDA to CDFT), ALDA to $f_{xc}(\mathbf{r},\mathbf{r}^*;\omega)$, and LDA to the static Kohn-Sham scheme.

in TDCDFT. By invoking the local density approximation to the tensorial xc kernel and exploiting this relation, we have constructed a new approximation form for $f_{xc}(\mathbf{r},\mathbf{r}';\omega)$. The resulting form adds a nonlocal nature to ALDA and the nonlocality is just enough to make the result for the friction coefficient of the electron gas free from the abovementioned contradiction in the vacuum limit.

In order to check the accuracy of this new form for $f_{xc}(\mathbf{r}, \mathbf{r}'; \omega)$, we have recalculated the friction coefficient of the homogeneous electron gas at various electron densities and compared the obtained results with the experimental ones. Note that our results for the homogeneous electron gas can be directly compared with the experimental ones in simple metals, in view of the fact that at low velocities the energy loss of ions in solids is due mainly to the stopping power of valence electrons.

In Fig. 1, an example of such comparisons is shown at the electron density as specified by $r_s=2.2$ (r_s : the conventional nondimensional density parameter) corresponding to aluminum with incident ions penetrating from its (111) surface. In the figure, we have also plotted the results in ALDA as well as those in LDA to the static Kohn-Sham scheme which amounts to the calculation without including any dynamical xc effects.

As can be seen in Fig. 1, experiment exhibits an oscillatory behavior in the friction coefficient with increasing the atomic number of incident ions. This behavior, which is attributed to the shell structure of valence electrons around the ion, is reproduced well in all the calculation schemes, but the best quantitative agreement with experiment is obtained by our new scheme (namely, LDA to CDFT) for $f_{xc}(\mathbf{r},\mathbf{r}';\omega)$.

We expect that our new approximation form $f_{xc}(\mathbf{r},\mathbf{r'};\omega)$ awaits various applications in a wide range of problems concerning dynamical processes in electronic systems.

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Universal Magnetization Minimum in One-Dimensional Spin Gap Systems under Magnetic Field

Oshikawa Group

Spin gap systems have been vigorously studied both experimentally and theoretically over decades. Spin gap systems generally lack usual magnetic order even at zero temperature, thanks to quantum fluctuation. Thus they provide an ideal playground to study effects of quantum fluctuations. Various spin gap systems have been found experimentally, including one-dimensional ones such as Haldane gap systems and two-leg spin ladders.

The spin gap can be closed by an applied magnetic



Fig. 1. Magnetization of the S = 1 Heisenberg antiferromagnetic chain (Haldane chain) as a function of temperature, for various values of applied magnetic field above the critical field. Each curve shows a minimum of magnetization at a finite temperature. Such a minimum is absent in the magnetization of a classical chain (shown in red). The temperature at the minimum increases, as the field increases.

field. At zero temperature, the system undergoes a quantum phase transition at the critical field where the gap closes. Above the critical field lies a gapless phase. In two and higher dimensions, the gapless phase at zero temperature has a long-range order of transverse magnetization. However, in one dimension, quantum fluctuations are so strong that the long-range order is absent even in the gapless phase. Instead, the gapless phase can be described as Tomonaga-Luttinger liquid, which is the universal theory for many gapless systems in one dimension. In fact, the S = 1/2 Heisenberg antiferromagnetic chain, which is gapless already at zero field, can be also described as the Tomonaga-Luttinger liquid.

This implies that, once the gap is closed by the applied field, a one-dimensional spin gap system would become essentially identical to intrinsically gapless systems, in the low-energy limit at zero temperature. However, if we consider finite-temperature properties, "remnants" of spin gap appear in various ways. We examined numerically the temperature dependence of the magnetization in the



Fig. 2. The crossover diagram on the field-temperature plane. In one dimension, phase transition is absent at finite temperature. However, there can be a crossover between different regimes. In the present case, as the temperature is increased at a constant field, the system crossovers into the "quantum critical regime" governed by the non-relativistic, parabolic dispersion of magnons.

S = 1 Haldane chain under magnetic field, using Quantum Monte Carlo simulation. Above the critical field, we find a minimum in the magnetization as a function of the temperature as in Figure 1. Such a minimum does not appear in the magnetization of the intrinsically gapless S = 1/2 Heisenberg chain. In fact, the magnetization minimum can be understood as a signature of crossover between the "quantum critical" regime governed by the nonrelativistic quantum critical point and the Tomonaga-Luttinger liquid regime, as in Figure 2. This is different from the similar magnetization minimum in three dimensional systems, which corresponds to Bose-Einsten condensation of magnons.

We have revealed that the magnetization minimum is a universal phenomenon for field-induced gapless phases slightly above the critical field. In particular, the temperature T_m at the minimum, the applied field H, and the critical field H_c obey the relation $T_m = x_0 (H - H_c)$ when H is close to H_c , where $x_0=0.76238\cdots$ is a universal constant.

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Pt/water Interface — towards a Full Understanding of the Electrochemistry—

Sugino Group

In our daily life, we utilize many electrochemical reactions, such as, electrolysis of water, fuel-cell reactions, corrosion, and plating/etching. These phenomena would be used to ultimately solve many problems related to environment, clean energy, and bio-membrane when the electrode/ solution interface could be completely controlled in the nano-meter scale. However, the interfacial phenomena are so complicated and controlling the interface is not easy,



Fig. 1. The electrode dynamics at Pt(111). ESM-FPMD simulation on the Volmer step $(H_3O^+ + e^- \rightarrow H(ad) + H_2O)$ of the hydrogen evolution reaction. The hydronium ion H_3O^+ at the contact releases a hydrogen atom toward the surface accompanying the discharging. To accelerate the simulation, negative bias of -0.7 V vs. the standard hydrogen electrode (SHE) was applied to the interface.



Fig. 2. The layer distribution function of the water molecules (oxygen site). With increasing negative bias, structure of the interfacial water molecules take a multiple changes, and finally takes a hydrophobic contact layer is formed -0.81 V vs. SHE.

raising increased demand for a comprehensive and practical interface theory. Our recent activity has been directed towards establishing such theory.

At the interface H^+ or OH^- ion is adsorbed accompanying an electron-transfer. This so-called redox reaction is influenced by *macroscopic factors* including the bias potential, pH, counter ion density, and the electric double layer as well as *macroscopic factors* such as local bond making/breaking with the electrode, tunneling/hopping of electrons and protons, and hydrogen-bond network. This is the essence of electrochemical reactions and relating the microscopic and macroscopic parts comprehensively is the target of the research. Lack of information regarding the primary obstacle to our understanding. In this context, we have started by simulating the interface of a bare metal surface and pure water under the finite bias condition.

To approach the problem we have previously developed the effective screening medium (ESM) based first-principles molecular dynamics (FPMD) method, which is able to apply a finite bias to interfaces consisting of the metal slab model and water molecules [1]. The scheme is applied to the interface under a negative bias condition (-0.7 V vs. the standard hydrogen electrode (SHE)) to find that an additionally introduced hydronium ion (H₃O⁺) is attracted to the Pt electrode and is adsorbed accompanying a discharging process within the simulation time (7 ps) [2]. See Figure 1 for the snapshots. This is the first simulation of the most fundamental electrochemical reaction, which is known as the Volmer step (H₃O⁺ + e⁻ \rightarrow H(ad) + H₂O) of the hydrogen evolution reaction, and suggests important roles of the structure and response of the water molecules.

The structure of the interfacial water is further investigated to clarify a change with the bias potential. The interfacial water molecules take a multiple step to change the structure from the "H-up" to the "H-down" as the water monolayer does on the surface, until a hydrophobic contact layer is formed [3]. The hydrophobic nature can be seen in the dramatically reduced oxygen atom density in the layer distribution function. (See the distribution at around 5 Å in Figure 2)

The simulation was too simplified to be realistic although suggestive. In the second step we are now trying to improve the ESM-FPMD to incorporate the pH and counter ion effect as well as the tunneling of the electrons. Algorithms to simplify the simulation is also undertaken to overcome the limitations in the computational cell size and the simulation time. Once these improvements are done, we will then be able to greatly advance our understanding of the electrochemistry which has attracted chemists over 230 years since the first discovery, expanding the field of the surface/interface science.

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Phase Diagram of Dissipative Double-Well Systems

Kato Group

Quantum tunneling between two states subject to dissipation has been one of central problems in both physics and chemistry for several decades. A number of studies on dissipative quantum systems have revealed that dissipation strongly affects the quantum nature of the system. For example, destruction of quantum superpositions has been studied extensively in a dissipative two-state model (a spin-Boson model) [1], which is obtained by truncation from a double-well system. Further, in the presence of strong dissipation, quantum tunneling between two states is completely suppressed, and a particle is localized in one well.

Truncation to a two-state model is justified in the large barrier limit ($g = \hbar \omega / V <<1$), where V is a barrier height and ω is a frequency of small oscillation around the minima. If the barrier height V is comparable to $\hbar \omega$, the doublewell system is never mapped to a two-state model. Do the characteristic features of the dissipative two-state systems (*e.g.* decoherence, localization, etc.) survive in the presence

Fig. 1. Phase diagram of dissipative double-well systems. The dimensionless parameter $g = \hbar \omega / V$ expresses strength of quantum fluctuation, and α is a dissipation strength. The full square shows the phase transition determined by our simulation with an accuracy of less than 1%. The full circle shows the coherent-incoherent crossover determined from the dynamical structure factor, where their statistical errors are typically given by the symbol size. The empty square and circle show the known transition and the crossover point in the large-barrier limit, respectively.

of large quantum fluctuation? We have studied this problem by using the path-integral Monte Carlo (PIMC) method [2, 3].

The conventional PIMC method applied to dissipative double-well systems has several difficulties. Most difficult problem is that the simulation time is proportional to $L^2\log L$, where L is a Trotter number (the division number of the path along the imaginary time). This makes it hard to treat the low-temperature region corresponding to a large L. We have developed a new algorithm of the PIMC for efficient local update. Our idea is based on a decomposition of the weight of the nonlinear potential term to several Gaussian distributions. In our algorithm, the simulation time is proportional to LlogL. Our method also reduces the correlation time along the Monte Carlo step. For example, to obtain 1000 independent samples for $L=2^{14}$ on a standard desktop personal computer, it takes about one month in the conventional method, while only 5 minutes in our improved method. To overcome critical slowing down, we also introduced cluster algorithm proposed by Werner and Troyer [4].

The localization transition occurs at α =1 in the two-state limit ($g \rightarrow 0$), where α is a dimensionless dissipation strength. This belongs to the Kosterlitz-Thouless transition [1]. Also for a finite g, it is expected that the transition belongs to the same universality class because of symmetry of the model. Therefore, we can analyze the transition by a finite-size scaling on the helicity modulus $\Psi = (\alpha/2N)\Sigma_{i,j} \langle x_i x_j \rangle$, where x_i is a discretised path. The upper curve of Fig. 1 shows the localization transition line determined by the PIMC simulation up to $L=2^{14}$. The coherent-incoherent crossover is defined in several ways. Here, we determine it by disappearance of a peak in the structure factor $S(\omega)$, which is obtained from the correlation function with the Padé approximation. The lower curve of Fig. 1 shows the crossover line.

As seen in the phase diagram, both the crossover and transition line shift towards a large α for a finite *g*. Furthermore, the tendency of saturation is clearly observed for larger values of *g*. These behaviors indicate that even for the infinitesimal potential barrier ($V \ll \hbar \omega$), a finite strength of dissipation is sufficient for decoherence and localization. Our results also indicate that the basic phenomena discussed in the dissipative two-state system can be observed also in the general dissipative double-well systems even for a small potential barrier.

Our improved algorithm can be applied to other problems with continuous variables. One typical example is the ϕ^4 chain. Our idea of Gaussian decomposition suggests that such a model can be mapped approximately into another model only with discrete variables after integrating out the continuous variables. This may give a new insight into the original model.

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Stripe Phase of Two-Dimensional Hole Gas at Half-Filled Higher Landau Levels

Iye Group

When a two-dimensional electron system (2DES) is subjected to a strong perpendicular magnetic field, electrons execute cyclotron orbital motion. Quantum mechanically, the cyclotron radii are quantized to discrete values and the corresponding energy spectrum is a series of discrete Landau levels (LLs). In such a situation the effect of Coulomb interaction becomes particularly crucial. This is reflected to the rich variety of the ground states of 2DES under a quantizing magnetic field depending on the LL filling factor v = nh/eB. The strongly correlation gives rise to fractional quantum Hall liquid states and Wigner crystal states at lower LL fillings. At half-filling of higher LLs, such as v = 11/2, 9/2, and 7/2, the 2DES was found to develop outstanding anisotropy in resisitivity in spite of no obvious source of anisotropy [1]. This was subsequently called stripe phase, which is a sort of charge density wave state, namely spontaneous development of spatial regions with different LL fillings. The stripe phase is sensitively affected by an in-plane magnetic field [1] or by an artificial potential modulation [2].

The previous studies were mostly focused on the 2D electron systems. Comparative studies on two-dimensional hole system (2DHS) having different parameter values from 2DES (*e.g.* higher effective mass) should shed light on the physical essence of the phenomenon. Although there were a few studies addressing to the stripe phase in 2DHS, the

Fig. 1 (a) Magnetoresistance traces of a 2DHS sample depicted in the inset. The two traces represent resistivity along two different current directions. (b) Temperature dependence of the peak resistivity at v = 7/2 for the two directions. The green data represent their ratio.

samples used in those studies were grown on the (113)A surface of GaAs substrate with Si dopant, so that they had inherent crystallographic anisotropy in the 2DHS plane. In the present work, we used 2DHS samples grown on the (001) substrate with Be dopant [3]. Figure 1(a) shows magneto resistance of a 2DHS sample for two different current directions. The QH transition peaks exhibit distinct anisotropy at half-filling of higher LLs, such as v = 11/2, 9/2, and 7/2 (but not at v = 5/2), indicating the occurrence of stripe phase at these fillings. Figure 1(b) shows the temperature dependence of the peak height at v = 7/2 revealing that the stripe phase disappears at around 150 mK. The effect of an in-plane magnetic field on the anisotropy is consistent with the behavior of the stripe phase elucidated for 2DES. However, the effect is found to be weaker than the case of 2DES, suggesting a stronger orientational pinning mechanism for the hole stripe to the underlying crystalline structure.

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Quantum Decoherence in Superconducting Charge Qubit

Katsumoto Group

Quantum decoherence is one of the biggest mysteries in quantum mechanics. Several mechanisms have been proposed for a system to decohere. Most interesting is the one that predicts a "phase transition" between quantum mechanical region and classical region. Superconductorinsulator transitions in Josephson array systems were claimed to be indirect evidences of the mechanism.

Fig. 1. Color plot of the source-drain current I_{sd} through the Cooper pair box against the plane of the gate voltage V_g and the source-drain voltage V_{sd} .

Fig. 2. Applied microwave frequency as a function of split PAJQP peak position measured from the original JQP peak. Blue and red circles correspond to the cases with and without two-dimensional electrons.

However such transitions are consequences of so complicated many-body interactions and far from simple, sound examples. Successful observation of coherence oscillation of charge qubit in a Josephson junction system was benefited from lacking of kinetic freedom in the superconducting energy gap. This situation, which is an ideal stage for quantum information processing, at the same time can be a test stage of quantum decoherence mechanism. Here we report tuning of quantum decoherence through the environment kinetic freedom.

The energy gap region is like a blank stage with qubit as the single player. The qubit in the present experiment is composed of a "single Cooper pair box", two Cooper pair reservoirs (source and drain) and two Josephson junctions, which connect these each reservoir and the box. In the single Cooper pair box, the number of Cooper pairs is quantized with the Coulomb energy. One of the Josephson junctions is composed as an interferometer so that the Josephson energy can be tuned by external magnetic field. All of these structures are made of vacuum evaporated aluminum. Now we introduce the other players. The qubit device is placed on an AlGaAs/GaAs heterostructure, to which two dimensional carriers can be introduced by light illumination. Such free carriers interact with Cooper pairs in qubit through Coulomb interaction. With increasing the number of electrons, the number of kinetic freedom increases and at the critical point the transition to classical region is expected.

The quantum coherence can be most easily and clearly observed in the frequency domain, where energy level anti-crossing due to the coherence between two different charge states is observed. For such experiment, we utilized current peaks due to Josephson-quasiparticle (JQP) tunneling, in which a Cooper pair tunnels through one of the junctions and two quasi-particles tunnel through the other. Figure 1 shows color plot of the current through the device as a function of the gate voltage and the sourcedrain voltage. White colored high current regions are the JQP peaks. With applying microwave, these peaks split into two or three due to the photon-assisted (PA) JQP process. The PAJQP process is caused by microwave excitation from lower energy level to upper thus a kind of dispersion relation is obtained from the microwave frequency dependence of the split width.

Figure 2 shows the microwave frequency as a function of split width for the device with and without two-dimensional electron system. Apparently the anti-crossing width is diminished for the device with two-dimensional electrons and the result evidences quantum decoherence due to the interaction between the Cooper pairs and the two-dimensional electrons.

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Superstructure-dependent Decoherence of Surface Electrons on Ge (001)

Komori Group

Electron loses its phase memory when it is inelastically scattered. The way of decoherence depends on the scattering mechanism. On metal and semiconductor surfaces, the observed decay of electronic standing waves is attributed to the decoherence, and is determined by electron-phonon or electron-electron scattering. We have studied the decoherence of the surface state electrons at the clean Ge(001) surface, where two stable superstructures with almost the same quasi one-dimensional (1D) electronic state coexist at 80 K.

The surface consists of buckled Ge dimers, which align in the [110] direction and form dimer rows. The empty π^* -band made of the dangling bond states of the surface Ge dimers behaves like a 1D free electron along the dimer row. There are two stable superstructures, $p(2\times2)$ and $c(4\times2)$ (see Fig. 1), and we can control the surface structure reversibly between them with hysteresis by changing the sample bias voltage V_B for the surface observation using scanning tunneling microscopy (STM). [1] We measured the decay of the π^* -band standing waves from monoatomic steps in

Fig. 1. Topographic (a) and dI/dV (b) images of a clean Ge(001) surface at 80 K. Two superstructures, $c(4\times2)$ and $p(2\times2)$ shown schematically in the upper panels, were intentionally positioned at the lower terrace of the step by a surface manipulation [1]. Standing waves of the π^* electrons from the step edge are seen in (b).

Fig. 2. Cross sections of the standing waves in the dI/dV images for c(2×2) (a,c) and p(4×2) (b,d) areas. [1] The sample bias voltage V_B is 0.5 V (a,b) or 0.4 V (c,d). The wave length decreases with increasing V_B. Blue and red curves are damping oscillation curves fitted to the data. The decay length of the wave for the same bias voltage is shorter on the $c(4\times 2)$ surface than on the $p(2\times 2)$ surface.

differential conductance dI/dV images after preparing a surface with the both superstructures by the $V_{\rm B}$ change.

Figure 1(a,b) shows the topographic and dI/dV images of the same area. On this surface, both $p(2\times 2)$ and $c(4\times 2)$ structures coexist on the lower terrace of a single step [1]. We can see that the sanding waves from the step decay in several nm in Fig. 1 (b). The cross sections of the differential conductance image from the step are plotted in Fig. 2 for $V_{\rm B} = 0.4$ and 0.5 V on the p(2×2) and c(4×2) areas. We fitted each standing wave to the equation: dI/dV(x) = $Aexp(-2x/L_c)sin(2kx+\delta)$. Here the fitting parameter L_c is the decay length of the wave, A and δ the amplitude and phase shift of the wave, and k the wave number of the π^* electron. The dispersion relation of the π^* -band for each surface obtained by this analysis is almost common to the both surfaces. Whereas, L_c on the p(2×2) surface is longer than on the $c(4\times 2)$ for both bias voltages. This suggests that the superstructure-dependent electron-phonon interaction in the two surfaces makes the decoherence of the surface electrons different.

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Microscopic diffusion processes of NO on the Pt(997) surface

Yoshinobu Group

The microscopic diffusion processes of NO molecules on Pt(997) at low coverage were investigated using time-resolved infrared reflection absorption spectroscopy (TR-ITAS). When NO molecules absorb on the Pt(997) surface at low temperature, each molecule transiently

Fig. 1. IRAS spectra of NO on Pt(997) as a function of substrate temperature. The coverage of NO is about 0.08 ML.

migrates on the surface from the first impact point to a possible adsorption site. At 11 K, the molecules are trapped at four adsorption sites on Pt(997): the on-top sites on the (111) terrace (OT), the hollow sites on the (111) terrace (HT), the bridge sites at the step (BS) and the hollow sites at the step downstream (HS), which have been evidenced by infrared reflection absorption spectroscopy (IRAS) (Figure 1) [1] and first-principles calculations [2].

Fig. 2. (a) A series of TR-IRAS spectra of NO on Pt(997) after an NO gas injection. (Coverage: 0.02 ML, Temperature: 76 K), (b) The change of the fractional coverage of each NO species in Figure 2(a) as a function of the elapsed time. Dots: OT species, cross: HT species, triangles: BS species, lines: fitted lines based on the first order kinetics model.

Fig. 3 Probable diffusion routes to the bridge site of the upper step. Route A: from the upper terrace, Route B: from the lower terrace. The starting point is set at an on-top site in this figure as an example. *: These adsorption energies are taken from Ref. [2].

By heating the surface to 45 K, the HS species migrate up to the BS sites; the migration barrier is roughly estimated to be 120 meV. In the temperature range from 70 K to 77 K, TR-IRAS measurements were carried out to observe the site change of OT species to the adjacent HT sites at isothermal conditions (Figure 2). Based on the Arrhenius analysis of time-resolved IRAS data at several temperatures, the activation barrier and the pre-exponential factor are estimated to be 200 meV and 2×10^{11} s⁻¹, respectively.

In the temperature range from 100 K to 110 K, the HT species migrate across the terrace and finally reach the BS sites. The activation barrier between the HT sites and its pre-exponential factor are estimated to be 290 meV and 6.5×10^{11} s⁻¹, respectively, form the TR-IRAS data together with kinetic Monte Carlo simulations.

On the whole, the quantitative microscopic picture of NO migration on Pt(997) has been established. Figure 3 summarizes the schematic diagram of NO on Pt(997) and the potential energy surfaces across the possible migration routes.

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Superconductivity of Nano-size Island Structures Studied by Scanning Tunneling Microscopy/ Spectroscopy

Hasegawa Group

Superconductors whose size are comparable to as small as their coherence length exhibit properties quite different from those of bulk counterparts. In order to study them in real space, we first prepared nanosize flat-top single-crystalline Pb island structures on a Si substrate, whose typical sizes are 25-75 nm in radius and 3 nm in thickness, and then investigated their superconducting properties and size effects *in situ* using a low-temperature scanning tunneling microscopy / spectroscopy.

Figure 1(a) show a 9 monolayer-thickness (ML) Pb island whose size is a few times larger than the coherent length (ξ). Tunneling spectra showing a superconducting gap were taken on this island under various magnetic fields perpendicular to the surface plane. It turned out that the gap, becoming shallow with the magnetic field [1,2], shows spatial dependence in the island.

In order to elucidate the observed site dependence, we made an image of the tunneling conductance at the bottom of the gap (zero bias conductance; ZBC) The ZBC image taken at zero field (b) indicates homogenous superconductivity. At 0.5 T (c), however, ZBC around the peripheral region is a little bit larger than the center due to the field penetration. Then, at 0.6 T (d), only the center region has high ZBC due to a vortex formation, which was confirmed by an agreement of its profile with the result of linearlized Ginzburg-Landau (LGL) equations (f). Finally, at 0.9 T (e) all area has high ZBC, indicating complete breaking of superconductivity in all the area.

Visualization of the vortex formation and the breakdown of superconductivity by STM/STS enables us to determine critical fields of the vortex penetration/expulsion (H_{p1}/H_{ex1}) and the complete breakdown of superconductivity (H_{SN}). We measured the critical fields on various sizes of islands. The results taken on 9ML islands are plotted in Figure 2. It turned out that there is a critical island size for the vortex formation. Larger islands tend to have lower vortex penetration/expulsion and breakdown fields. According to theoretical studies based on LGL equations, the islands whose radius is smaller than 1.3 times coherent length do not form

Fig. 1. (a) STM image of a 9ML Pb island showing an area where the following ZBC images were taken. (b)-(e) ZBC images taken under the magnetic field of 0.0 T (b), 0.5 T (c), 0.6 T (d) and 0.9 T (e). (f) cross-sectional ZBC profiles. A line drawn for the 0.6 T data is a spatial profile of the order parameter calculated from the LGL equations.

Fig. 2. Lateral size dependence of various critical fields; the transition field between the superconducting and normal states H_{SN} , vortex penetration H_{p1} and vortex expulsion H_{ex1} . All critical fields were obtained from 9ML-thickness Pb islands.

vortices. From the critical size, the coherence length of the 9ML islands was estimated to be 31+3 nm. As the radius increases H_{SN} approaches to that of thin films; namely, the critical field for the surface superconductivity H_{c3} , which is given by $1.695 \times h/4\pi e\xi^2$. The value of H_{c3} calculated from the estimated coherence length is marked in the plot. Although this analysis neglects an effect of island shape, the experimental results are quite consistent qualitatively with the results of the theoretical studies.

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Mapping Defect States in SrTiO₃

Lippmaa Group

Despite the seemingly simple crystal structure, the electronic properties of SrTiO₃ continue offer surprises, such as metallic and even superconducting interfaces in SrTiO₃ heterostructures. SrTiO₃ is an unusual material in the sense that transitions from the band insulating state to metallic and even superconducting states occur at very low carrier densities, at around 10¹⁸ and 10²⁰ cm⁻³, respectively. While this feature suggests several possible avenues for doping experiments, such as charge transfer or field effect, it also means that SrTiO₃ is prone to inadvertent doping by a very low density of defects, such as oxygen vacancies. Since the Fermi level in intrinsic SrTiO₃ is pinned very close to the conduction band edge, it is more accurate to view SrTiO₃ as a wide-gap semiconductor ($E_q = 3.2$ eV), rather than a classical band insulator. In intrinsic SrTiO₃, the Fermi level is about 100 meV below the conduction band (CB) bottom. In a SrTiO₃-based field-effect transistor

Fig. 1. The band structure of $SrTiO_3$ close to the conduction band (CB) bottom. The intrinsic Fermi level is located about 100 eV below the CB edge. In a field-effect transistor (Inset) the Fermi level of $SrTiO_3$ can be modulated by applying a positive or negative bias to the gate electrode.

(Fig. 1), the Fermi level can be pushed higher or lower by accumulating or depleting carriers from the device channel. The distance between the Fermi level and the CB edge can be measured by determining the activation energy for thermally activated conduction in the transistor channel.

The temperature dependence of channel current in a partially oxygen-depleted $SrTiO_3$ device is shown in Fig. 2. Particularly in the depletion regime, where the gate bias is negative, the channel current shows a strong temperature dependence. By measuring the channel current as a function of temperature at a fixed gate bias, the activation energy can be found from an Arrhenius plot. Since the number of carriers depleted or accumulated in the FET channel (and thus the position of the Fermi level) is a linear function of the gate bias and gate insulator capacitance, it is possible to estimate the density of states as a function of energy (from the CB bottom).

The plot in Fig. 3 shows a linear increase in E_A when the gate bias is close to zero, corresponding to approximately

Fig. 2. Channel current of a $SrTiO_3$ FET as a function of gate bias and temperature. In depletion mode (negative gate bias) the channel current is a strong function of temperature due to thermally activated transport in localized in-gap states close to the conduction band edge.

Fig. 3. Activation energies determined from the temperature dependence of channel current for each gate bias. At $V_g = +0.5$ V, the device becomes metallic, i.e. the Fermi level is pushed across the mobility edge. For negative bias, the activation energy increases linearly (constant density of states) until saturating at 0.3 eV, which corresponds to the start of an in-gap impurity band related to oxygen vacancies in SrTiO₃.

constant density of states. An abrupt saturating behavior is seen below about -0.5 V, where the activation energy does not increase beyond 0.3 eV. This shows that in oxygen-deficient SrTiO₃, there is a large density of states at about 0.3 eV below the CB bottom. We assign this state to ogygen vacancies in SrTiO₃.

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Quantum Transport Simulations of Carbon Nanotube Field-Effect Transistors Using O(N) Time-Dependent Approach

Kobayashi Group

Toward developments of nanoscale devices, intensive works have been performed in the investigation of the transport properties of nanoscale systems. Carbon nanotubes show various electronic states and are expected to be a candidate element of nanoscale devices. We have investigated the quantum transport properties of carbon nanotube field-effect transistors using time-dependent wave-packet diffusion method [1,2]. We have succeeded in O(N) calculations for transport of large systems with micronorder lengths using an atomistic model and investigation from ballistic regime to diffusive regimes based on the Kubo-Greenwood formula.

We have investigated the effects of electrode contact and electron-phonon coupling on the quantum transport properties of carbon nanotube devices with micronorder channel lengths. We have elucidated the electron mobility of a (10,0)-semiconducting carbon nanotube fieldeffect transistor with several μ m channel lengths for the

Fig. 1. O(N) calculation time of the conductance of the (5,5) carbon nanotubes with various system lengths on a Hitachi SR11000 in ISSP. The solid and broken lines represent the calculation time of the Chebyshev polynomial development and the split operator methods, respectively. In the inset, we show the calculation time for the short system lengths [2].

Fig. 2. Calculated time-dependent diffusion coefficient of the (10,0) carbon nanotube field-effect transistor with 1 μ m channel length for various gate voltages V_g [1].

applied gate voltages in the absence of phonons, vacancies, and impurities. Furthermore, we investigated the phonon scattering effects on the mobility and conductance of the carbon nanotubes. The longitudinal acoustic phonon and optical phonon modes with a small momentum were considered at room temperature. The conductance of a (5,5) metallic carbon nanotube is decreased around the Fermi energy by the optical phonon scattering. On the other hand, the mobility and conductance of the (10,0)-semiconducting carbon nanotube are decreased by the acoustic phonon mode. We have also elucidated the transitions of transport behaviors of nanotubes from ballistic to diffusive regimes.

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Two Dimensional Anti-ferromagnetic Solid ³He in High Magnetic Fields

Ishimoto Group

³He film adsorbed on graphite surface is one of the ideal two-dimensional (2D) Fermi systems that 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 Mott transition [1]. The 2D solid form 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 two-particle exchange play an important role. The exchange of even number of particles is anti-ferromagnetic (AFM), while that of odd ones is ferromagnetic (FM). The competition among them, in addition to the geometrical frustration inherent to a triangular lattice structure, makes AFH solid ³He film strongly frustrated.

According to recent theoretical studies in the exact

Fig. 1. Temperature dependence of magnetization in the AFM 4/7 phase for various fields. The vertical axis is normalized by the saturation magnetization. The lines in the inset are the Brillouin functions.

Fig. 2. Magnetization curve for the AFM 4/7 phase at 0.7 mK. The vertical axis is normalized by the saturation magnetization.

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 interaction parameters obtained at high temperatures for the AFM phase (so called a 4/7 phase) suggests a quantum spin liquid (QSL) ground state with a finite excitation gap [2], and a magnetization plateau. Experimentally the heat capacity and susceptibility measurements indicate a gapless spin liquid ground state [3][4]. Our previous measurement with a double–gradient Faraday-type magnetometer suggests the existence of magnetization plateau in the AFM 4/7 phase on ⁴He mono layer pre-plated graphite. To resolve such a discrepancy, we investigated the magnetization curve by using NMR technique over the wide frequency range from 7 MHz to 360 MHz in a single experimental run.

Figure 1 shows the magnetization of AFM 4/7 phase on ⁴He mono layer pre-plated graphite as a function of temperature for various magnetic fields. Here the vertical axis is the polarization estimated from the saturation magnetization obtained by fitting the temperature dependence in the high temperature region. Figure 2 shows the magnetization curve obtained from Fig.1. It shows a clear plateau at half of saturation magnetization, followed by its full saturation at about 10 T. It is essential to take account of five and six multiple spin exchange interactions to explain the results.

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First Evidences of Vortex Fluid to Supersolid Transition in Solid ⁴He

Kubota Group

The question, whether particles forming a solid lattice, can show also properties of superfluid at the same time, has a long history. From the first theoretical study's negative answer [1] to positive discussions [2], the original idea of possible superfluidity was discussed based on Bose-Einstein condensation(BEC) of particles themselves or imperfections in the system of a quantum solid. The latter refers to either vacancies or interstitials and dislocations or all together, but in practice only the simplest vacancies and interstitials had been evaluated and from estimated densities of imperfections in solid ⁴He the occurrence of such a state, now called "Supersolid" had been expected to be at very low temperatures. The situation has been changed drastically since the discovery of so-called "non-classical rotational inertia" (NCRI) in solid ⁴He, by Kim and Chan [3]. NCRI is common in all known other superfluids and it had been suggested to be observable by Leggett[2-c] long time ago for a supersolid.

It then became a problem since reported "onset temperature, T_0 " was higher than 150 mK or 300 mK, and not much different (within a factor of 10) from liquid ⁴He λ line (2.17-1.743K, depending on pressure), where superfluid transition is primarily governed by BEC of ⁴He atoms. P. W. Anderson [4], then proposed that the reported "NCRI" in solid He is actually "non-linear rotational susceptibility (NLRS)" of

(a)

Fig. 1. (a) TO Energy dissipation change ΔQ^{-1} vs DC angular velocity Ω . Extra dissipation appears over a temperature range T < T_c 75-85 mK below a dissipation peak (round peak) and the linear dependence to Ω at constant temperatures is more clearly seen in (b) and it gives clear evidence of vortex lines penetration, in contrast to a possible Ω^2 dependence expected to be caused by solid vibrations or other solid properties under DC rotation.

vortex fluid state and he pointed out that real T c of supersolid transition should exist at some lower temperature.

Due to the very stable torsional oscillator conditions [5] and the world leading rotation cryostat [6], which was constructed at the ISSP, we are able to report and confirm the following points:1). From detailed AC velocity dependent responses of torsional oscillator (TO) containing solid ⁴He samples we could determine uniquely an onset temperature T_o, which is much higher than so far reported data. We discuss the appearance of vortex fluid state below T_0 [7]. 2). An indication of transition from vortex fluid state to the real 3D supersolid state is discovered below $T_c = ~75 \text{ mK}$ [8]. 3). Existence of vortex lines though the solid ⁴He sample under DC rotation below T_c is confirmed by the linear dependence on DC rotational speed Ω , Fig.1(b) [9]. We can comment on possible misinterpretation in the recent report by Day and Beamish on the "hardening" of the Solid ⁴He [10]. For recent reviews we find [11, 12].

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Interlayer Magnetotransport in Layered Conductors: **Multilayer Dirac Electron Systems** and Magnetic Breakdown Systems

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Interlayer magnetotransport in two multilayer electron systems has been investigated experimentally and theoretically.

(1) Dirac electron systems

We have considered the interlayer magnetotransport in multilayer Dirac electron systems, in which two-dimensional electron layers with Dirac cone dispersion stack with weak interlayer coupling. The Fermi level is assumed to be at the contact point of Dirac cones, so that the systems become zero-gap conductors. Under magnetic fields, the n=0 Landau level always appears at the contact point (zeromode), and its overlap with other Landau levels vanishes at rather weak field. In this quantum limit, the interlayer transport is dominated by the tunneling between zero-modes on neighboring two layers. The increase of zero-mode degeneracy causes strong negative interlayer magnetoresistance under normal magnetic fields. Spin splitting of the zero-mode could change the negative magnetoresistance to positive in high magnetic fields and low temperatures.

When parallel component of magnetic field is additionally applied, the interlayer resistance recovers from negative magnetoresistance reflecting the decrease of overlap of two zero-modes between which tunneling is allowed. The present model has successfully explained the magnetotransport features observed in an organic conductor α -(BEDT-TTF)₂I₃ by Tajima and Kajita. This fact strongly supports the realization of a Dirac electron system in α -(BEDT-TTF)₂I₃ predicted theoretically.

Fig. 1. (a) Dirac cone dispersion and (b) Landau level configuration in the 2D massless Dirac electron system. (c) Interlayer conductivity in the multilayer Dirac electron system in magnetic fields calculated using a lattice parameter of α -(BEDT-TTF)₂I₃.

Fig. 2. Second derivative pattern of interlayer resistance of α -(BEDT-TTF)₂KHg(SCN)₄ scanned over the whole magnetic field orientation.

(2) magnetic breakdown systems

We have studied the change of angle-dependent magnetoresistance oscillation phenomena through magnetic breakdown process. Using the double-axis rotator, we have measured the angle-dependent pattern of interlayer magnetoresistance of an organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ over whole magnetic field orientation as a function of temperature, and obtained complicated mixture of various angular effects. The electronic system of CDW phase of α -(BEDT-TTF)₂KHg(SCN)₄ can be considered as the quasi-two-dimensional (Q2D) electron system with cylindrical Fermi surface under additional CDW potential. The CDW amplitude is controlled by temperature. Using quantum tunneling picture, we have shown that the Kajita oscillation of the original cylindrical Fermi surface is modified by the Lebed-like resonance with decreasing temperature, since the magnetic breakdown through the CDW gap is suppressed. The present model has well explained the observed features in α -(BEDT-TTF)₂KHg(SCN)₄.

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Effect of Pressure on the One-dimensional Organic Conductor (TMTTF)₂SbF₆

Uwatoko Group

The $(TMTCF)_2X$ series (C = S and Se, X = monovalent anion) comprises traditional quasi-one-dimensional compounds exhibiting highly anisotropic electron transfers. Almost two decades ago, a general pressure temperature (P-T) phase diagram was proposed for (TMTCF)₂X families. The compound (TMTTF)₂SbF₆ has the largest monovalent anion in the (TMTTF)₂X series, which is advantageous to investigate the wide range of the P-T phase diagram for a single system. The ground state of this compound has an AF order. The transition temperature is $T_{\rm N} = 8$ K and a charge order (CO) occurs at $T_{\rm CO} = 154$ K. Combining these facts with the concepts embodied in the general phase diagram, the possible sP and SDW phases exist in (TMTTF)₂SbF₆ at low pressures, and the SC phase may be stabilized with further application of pressure. Therefore, the systematic investigation of (TMTTF)₂SbF₆ has been eagerly anticipated in order to understand the entire electron correlation system in all the (TMTCF)₂X salts.

We obtain the general P-T phase diagram by adding (TMTTF)₂SbF₆, as shown in Figure 1. In the figure, the shift of pressure axis in (TMTTF)₂PF₆ and (TMTTF)₂AsF₆ are estimated to occur at 1 GPa and 0.5 GPa, as indicated by the arrow. The possible sP, AF, and SDW regions are referred to the previous general (TMTCF)₂X phase diagram. After the CO and AF phases vanish in (TMTTF)₂SbF₆, new phase appears in resistivity. The transition temperature $T_{\rm S} \sim 20$ K of the SbF₆ salt is comparable to the temperature of the sP and SDW transitions in the AsF₆ and PF₆ salts. A further increase in the pressure probably reduces the effective on-site Coulomb interaction and breaks the nesting of the Fermi surface. Then, the superconductivity appears at 5.7 GPa, finally. The SC phase is observed over the wide pressure range of 5.4 < $P \le 9$ GPa. As compared to the SC phase of other TMTTF compounds, the pressure range is anomalously wide. The SC region of PF₆ and AsF₆

Fig. 1. P-T phase diagram of $(TMTTF)_2SbF_6$ constructed from the data obtained in this study. It is superposed on previously obtained phase diagrams of $(TMTTF)_2FF_6$ and $(TMTTF)_2AsF_6$. The notations of CO, AF, sP, SDW, and SC refer to charge order, antiferromagnet, spin-Peierls, spin-density-wave, and superconducting states, respectively. The data obtained in this study are described as solid green diamonds (T_P), as solid light blue circles (T_S), and solid blue circles (T_C). The SC regions of the PF₆ and AsF₆ salts are drawn in the SC phase of SbF₆.

are described as a violet shadow in Figure 1. Moreover, the SC phase of $(TMTTF)_2SbF_6$ has two local maxima and the SC phase is deeply into the T_S phase. With regard to the double maxima in the SC phase, the heavy fermion materials CeCu₂Si₂/Ge₂ have been known to depict this feature. It could be possible that the mechanism underlying the formation of the double maxima in $(TMTTF)_2SbF_6$ is similar to that in heavy fermion compounds. To clarify the superconducting properties, more detailed investigations are currently underway.

From the previous reports of the (TMTTF)₂X family, the AF ground state (commensurate SDW) appears between the sP and the SDW states. The AF state corresponds to the ground state of (TMTTF)₂Br at the ambient pressure $(T_{\rm N} = 15 \text{ K})$. Based on previously obtained phase diagrams, an unusual ground state change such as AF(1)-sP-AF(2)-SDW-SC can be expected in (TMTTF)₂SbF₆, but the expectation is not consistent with dimensional effect by pressure. Recently, ¹³C-NMR and ESR measurements indicated that the characteristic features of the AF state at the ambient pressure in (TMTTF)₂SbF₆ (AF(1)) and (TMTTF)₂Br (that is, AF(2)) are slightly different. While SbF_6 has considerable 3-D AF interaction, the nature of AF(2) in (TMTTF)₂Br is characteristic of a low-dimensional magnet. Furthermore, the AF wave vector Q = (1/2, 0, 0) for (TMTTF)₂SbF₆ is different from that (Q = (1/2, 1/4, 0)) for $(TMTTF)_2Br$. It is natural to consider that the differences in the nature of the AF phase between (TMTTF)₂SbF₆ and (TMTTF)₂Br are related to the existence of a clear or nonclear CO phase in paramagnetic region. Therefore, we cannot deny the possibility of the existence of an AF phase like (TMTTF)2Br (AF(2)) after the disappearing the CO phase. And also, sP phase may exist between two AF phases in (TMTTF)₂SbF₆.

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Photo-induced Metallic State Observed by Terahertz Pump-probe Spectroscopy

Suemoto and Hiroi Groups

Recently, we have been studying the photo-induced phase transitions by using terahertz (THz) pump-probe spectroscopy. Here, we present the picosecond dynamics of the insulator-metal transition induced by photoexcitation in vanadium dioxide (VO₂) [1]. VO₂ exhibits an insulator to metal phase transition at 340 K accompanying a structure change from rutile to monoclinic phase. THz pulses, whose spectral range is from 0.4 to 12 meV, can probe directly the ultrafast free carrier response. The dynamical behavior after photoexcitation at 800 nm in VO₂ thin film was investigated by the optical-pump THz-probe spectroscopy.

Figure 1 shows the temperature dependences of the transmittance of the THz wave and the electrical resistivity in steady state. The decrease of the transmittance corresponds to a decrease of the electrical resistivity, that is, an increase of dc electric conductivity. The good agreement between the transmittance and the resistivity indicates that the THz probe pulses detect properly the free carrier response of the metallic state. The transmitted THz waveforms with optical pump excitation at different time delays are shown in Fig. 2. After the photoexcitation, the amplitudes of the THz waves decrease with increasing the delay time Δt . In order to investigate the THz response in detail, the time evolutions of the transmittance wave evalu-

Fig. 1. Temperature dependences of the power transmittance of the THz pulse (left axis) and the normalized electric resistivity (right axis).

Time (ps)

Fig. 2. The transmitted terahertz waveforms with the photoexcitation at different delay times between optical pump and THz probe pulses Δt .

Fig. 3. The temporal evolutions of the transmittance changes at different excitation densities.

ated as shown in Fig. 3. The temporal profiles show a drastic change around 4.0 mJ *i.e.*, the transmission change increases significantly above this excitation density. The decay rates increase from 500 ps to 20 ns with increasing the excitation density. These transient changes of the transmittance recover in the time interval of the laser pulses of 200 μ s.

The time evolution of the transmittance at 4.0 mJ/cm^2 in Fig. 3 shows a two-step decrease, a fast onset within 800 fs and a gradual growth after 2 ps. This behavior is very similar to the temporal profile of the fraction of the high temperature phase measured by means of the time resolved X-ray and electron diffraction spectroscopy [2]. This good agreement indicates that the time profile of the transient transmittance change, which reflects the conductivity change, definitely corresponds to the fraction of the high temperature phase accompanying the change of the lattice structure. Furthermore, this fact indicates that the photoinduced insulator-metal phase transition is of the Peierls type.

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Firefly Bioluminescence: Quantum yield and pH-sensitive Color Change

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Firefly bioluminescence [1] is known as one of the most efficient light-emission systems on the earth having wonderful color change mechanism with pH, and has attracted extensive interest by many scientists not only in the field of biology but also in chemistry, physics, electronics, and photonics. The frequently and widely cited properties of firefly bioluminescence with extremely high efficiency and pH-dependent emission-color change between yellow-green and red modeled as chemical equilibrium between two corresponding states have been based on a seminal report in 1960 [1], which measured a high

Fig. 1. Quantitative luminescence spectra of firefly bioluminescence (*Photinus pyralis*) for 2.98×10^{11} luciferin molecules at various pH values with Tris or GTA buffers. The vertical axis is scaled by an absolute number of emitted photons in a unit of eV⁻¹. Integrated area of each spectrum gives total number of emitted photons, or integrated quanta of luminescence.

quantum yield of 88±25%. However, necessity for more reliable and refined experiments has long been addressed, because the same group later found and commented that luciferin has optical isomers and only the D-form contributes to bioluminescence, and that the purity of D-luciferin used in their measurement was probably significantly degraded by racemization. In spite of this problem, no correction or confirmation had been made. A new quantumyield determination with pure D-luciferin was long overdue.

We quantified quantum yields and emission-color change of firefly (Photinus pyralis) bioluminescence for pH from 5.4 to 8.9 [2], using our recently developed totalphoton-flux spectrometer [3] for bio/chemiluminescence. We determined the highest quantum yield of 41.0±7.4% (1 standard deviation estimate, coverage factor k=1) at pH 8.5. This value is still higher than but closer to those of other bio/chemiluminescence systems such as 28% in cypridina (sea firefly), 17% in aequorin (jelly fish), 30% in bacteria, and 1.24% in luminol. Moreover, we measured quantitative bioluminescence spectra in the absolute number of all emitted photons, as shown in Fig. 1, and found that all the spectra were systematically and quantitatively decomposed into one pH-sensitive and two pH-insensitive Gaussian peaks, and that no intensity conversion between yellow-green and red emissions but mere intensity variation of the pH-sensitive Gaussian peak at 2.2 eV causes the changes in apparent emission colors. These cast serious doubts on long-standing pictures of bioluminescence-color determination by the pH equilibrium between yellow-green and red states.

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Development of a New Spin-Polarized Photoemission Spectrometer with High-Efficiency and High Energy-Resolution

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Spin-resolved photoemission is a powerful tool to investigate core-level and valence band electronic structures of materials. In principle, one can determine all the quantum numbers of electron with this experimental technique. Usually, however, the spin-polarized photoemission is, a time-consuming experiment and one had to trade energyresolution off with reasonable data acquisition time. In order to conquer this weakness, we have developed a new spin-polarized photoemission spectrometer employing the very low energy electron diffraction (VLEED) type spin-polarimeter [1] in which spin-dependent reflection of low energy electrons at a ferromagnetic target is utilized for the spin detection [2].

As shown in Fig. 1, the system consists of the spin-polarimeter and a high energy-resolution energy analyzer (PHOIBOS-150, SPECS GmbH). After energy analysis, electrons are transported to the scattering chamber by electrostatic lens and scattered by the ferromagnetic target. Spin-dependent reflectivity of the electrons by the target with different magnetizations is measured by the channeltron located just beside the exit of the lens. The magnetization of the target is changed by a small coil beside the target. Since the reflectivity of the low-energy electron is much higher than the back scattering probability of high-energy electrons used for the usual spin-polarimetrer (Mott detector), the efficiency of the VLEED polarimeter is expected to be an order of magnitude higher than the Mott detector.

Despite of its high-efficiency, the polarimeter did not become common, because of their vexatious complication of preparing the target, which was usually an iron single crystal film on Ag(001) surface. In addition the rapid deterioration of the iron target by oxidation with residual gases was the other serious problem. These disadvantage can be overcome by employing the Fe(001)p(1×1)-O film grown on MgO(001) crystal for the target [3]. Hence, we have realized

Fig. 2. Photoemission spectrum of secondary electron of Fe(001) film (a) and the asymmetry (b) caused by the spin-polarization.

the high-efficient spin-polarized photoemission spectrometer by utilizing the new target.

Figure 2(a) shows the photoemission spectrum of the secondary electrons of Fe(001) film measured with UV light of He discharge lamp (HeIa, hv=21.22eV). The asymmetry of the reflectivity caused by the spin polarization of the film was observed as shown in (b). The maximum asymmetry is about 15%. We compared the value of the asymmetry with previously reported spin-polarization (~50%) of Fe(001) film [5], and estimated the Sherman function, the power of spin analysis of this polarimeter as 0.29±0.01. Considering the reflectivity of the electron by the target (~0.12), the efficiency of the spin-polarimeter, i.e. figure of merit (FOM): S²I/I₀, was determined as 1.0×10^{-2} . This value is approximately 100 times higher than that of a usual Mott type spin-polarimeter.

In Fig. 3, we show the electron kinetic energy dependence of the FOM. The energy dependence is in good agreement with previously reported results of the FOM obtained by the reflectivity or absorption measurement of spin-polarized electron at the Fe(001)- $p(1\times1)$ -O target (see inset). As shown in the figure, the maximum FOM is at around electron kinetic energy of 6 eV. The good performance with lower kinetic energy of this detector helps to

Fig. 1. Schematic view of the spin-polarized photoemission spectrometer. Spin polarization is measured as the asymmetry of electron reflectivity in different target magnetization.

Fig. 3. Electron energy dependence of the figure of merit (FOM) of the spin-polarimeter. Inset is previously reported results taken from the Ref. 4.

achieve spin-polarized photoemission measurements with a high-energy resolution, because one can set the pass energy of the electron energy analyzer at low values. With newly developed spin-detector, we measured spin-resolved photoemission spectra with a relatively high-energy resolution (~60 meV), and those with much higher energy resolution can be realized in near future.

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Shake gel: Shear-induced gelation

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Complex fluids, such as colloids, surfactant systems, polymer solutions, liquid crystals, and their mixtures exhibit a variety of rheological properties, such as depletion flocculation, shear thinning, shear thickening, thixotropy, rheopexy, shear-induced sol-gel transition, etc. [1]. In order to elucidate the sophisticated rheological behaviors of shear-induced gelation, we report here microscopic observation of a "shake gel" consisting of polymer and nanoemulsion (NE; the droplet radius, $R \approx 16.5$ nm) undergoing sol-gel transition by shearing. Simultaneous measurements of rheology and small-angle neutron scattering (SANS), hereafter we call Rheo-SANS, were carried out under shear flow as well as during relaxation process after cessation of shear [2].

Figure 1 shows the contour map of the dilatancy factors for Dx/y's obtained at 25 °C, where x and y denote the concentrations of the NE solution with respect to the that of a stock solution and of 2-hydroxyethyl cellulose (HEC), respectively. Here, the dilatancy factor was defined by the

Fig. 1. Contour map of the dilatancy factor as a function of NE and HEC concentrations. The numbers in the figure indicate the relative dilatancy factors.

Fig. 2 Shear viscosity η changes of the shake gel, D50/04 as a function of the shear rate $\dot{\gamma}$. The changes of η during the first ascent process can be classified into four regions, (A) the Newtonian, (B) the shear-thinning, (C) the shear-thickening, and (D) the slipping regimes. η in the first-descent process and the second loop was completely the same without any hysteresis. The dotted curve with diamonds, the dotted curve with triangles, and the dashed curve denote η 's for 0.4 wt% HEC and 50 % NE, and the sum of the viscosities of the HEC and NE, respectively.

viscosity ratio after and before vigorous shaking. As shown in the figure, D50/04 has the largest viscosity ratio (the number in the figure). This clearly shows that viscosity thickening occurs in a narrow concentration range of HEC and NE.

Figure 2 shows the shear viscosity η variation of D50/04 at 25 °C during cyclic changes of the shear rate $\dot{\gamma}$. The dotted and dashed curves show η 's for the reference samples, i.e., 0.4 wt% HEC solution (dotted curve with triangles) and 50 wt% NE (dotted curve with diamonds), and the sum of the two viscosity curves (dashed curve), respectively. Both behaved as a Newtonian fluid and a shear thinning occurred for $\dot{y} = 10 \text{ s}^{-1}$. \dot{y} increased stepwise from 10 s^{-1} for 30 s to 10^3 s⁻¹ for 3 s (the ascent process). After reaching $\dot{y} = 10^3 \text{ s}^{-1}$, \dot{y} was stepwise decreased to 10^{-3} s^{-1} (the descent process). The same cycle was repeated several times. As shown in the figure, the changes of η in the first ascent process can be divided to four regimes, (A) the Newtonian, (B) the shear-thinning, (C) the shear-thickening, and (D) the slipping regimes. In general, A- and B-type behaviors are commonly observed in polymer solutions. However, the regime C is characteristic of this system. η of the descent process is markedly larger than that in the ascent process and finally it reached a value 300 times as large as that of the beginning at $\dot{\gamma} \approx 10^{-3} \text{ s}^{-1}$. As a result, a transient sol-gel transition was observed in the steady state. Rheo-SANS measurements were carried out on D50/04 at various steady shear rates, where a double-cylinder geometry was employed and the neutron beams were shed in both the radial and tangential directions. Interestingly, the SANS intensity patterns remained isotropic and had a distinct peak maximum around q = 0.14 nm⁻¹, irrespective of the shear rate. Here, q is the magnitude of the scattering vector. However, a significant increase in I(q) was observed exclusively in the low-q region by shearing, indicating an occurrence of shear-induced phase separation.

On the basis of the experimental results disclosed above, we propose a mechanism of the shear-induced sol-gel transition. The key issue is the coexistence of *equi-spaced* nano-droplets and long polymer chains in a solvent. The

Fig. 3 Deformation model of shake gel. (a) Un-sheared ($\gamma = 0$) and (b) low shear ($\gamma = 1$).

size of the polymer chains is comparable to the interdroplet distance. Figure 3 schematically shows the relative sizes of the droplets and the polymer chains. The droplets are dispersed in the solvent with the inter-particle distance of 43.9 nm due mainly to electrostatic repulsive interaction and play as fixed hooks to pin polymer chains, around which each polymer chain with the hydrodynamic radius, $R_{\rm h,HEC} \approx 41.9$ nm, is located due to a weak van der Waals interaction and/or hydrogen bonding. At a stationary state, the polymer chains are not percolated and are localized near one or a few droplets Fig. 3a. Therefore, a shear strain does not generate restoring force, and hence a simple dissipative flow occurs (region A in Fig. 2). By shearing, the polymer chains are stretched in the flow direction. If \dot{y} is not large enough, the polymer chains do not link neighboring droplets. As a result, a viscosity thinning is observed (region B in Fig. 2). When $\dot{\gamma}$ increases further, the polymer chains are attached to neighboring droplets and a bridge is formed. When this bridging occurs throughout the system, a percolation transition occurs, and a shear thickening is observed (region C in Fig. 2, and Fig. 3b). At a higher shear rate, however, slippage on the surface and/or disruption of polymer chains takes place. As a result, viscosity thinning is observed (region D in Fig. 2).

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Neutron Scattering and Thermodynamic Studies of Ion Gels

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Recently, it was found that a series of organic ionic compounds have melting points lower than room temperature and they are called "room temperature ionic liquids" or simply "ionic liquids". These compounds are remarked as green solvents utilizing their nonvolatility, noncombustibility, etc. "Ion gels" consist of network polymers and solvents of ionic liquids [1]. They form transparent, soft, and strong membranes with high ionic conductivity, attracting much attention in applications for various electrochemical devices, actuators, etc. We are interested in the basic physical properties of ion gels, especially in the glass transitions and related dynamics of ionic liquids and polymers. In this study, we have measured the heat capacity and quasielastic neutron scattering of ion gel PMMA/EMITFSI where PMMA is poly(methylmethacrylate) and EMITFSI is 1-ethyl-3-methylimidazolium bis(trifluoromethan esulfonyl)imide.

Figure 1 shows the heat capacity difference between PMMA/EMITFSI gels and bulk PMMA ($T_g = 380$ K) plotted as values per mole of ionic liquids (IL). The data were measured by homemade adiabatic calorimeters in our laboratory. The glass transitions appeared above 170 K depending on the mole fraction of ionic liquids x(IL). The fact that all of the heat capacity curves coincide with each other below T_g implies that the additivity of heat capacity is valid below T_g . Two overlapping glass transitions occurred for the samples with x(IL) of 0.1, 0.18 and 0.3; the lower-temperature glass transition is due to the freezing of IL while higher-temperature one due to the freezing of PMMA. These data indicate that the motions of IL and PMMA are still decoupled in the region x(IL) < 0.5 but coupled in the

Fig. 1. Molar heat capacities of ionic liquid EMITFSI in PMMA/ EMITFSI gels. The parameter x(IL) is the mole fraction of the ionic liquid.

Fig. 2. Dynamic structure factor of PMMA/EMITFSI [x(IL)=0.3]. The data were fitted well by two Lorentz functions as shown by the black curve. The blue curve denotes the broader Lorentz component.

region x(IL) > 0.5. It was also found that the plasticization of PMMA due to IL is drastic in the lower x(IL) region and T_g does not change much in the higher x(IL) region.

Figure 2 shows the dynamic structure factor of ion gel d-PMMA/EMITFSI, where PMMA was deuterated to extract the motion of ionic liquids with many H atoms; the neutron scattering cross section of H atom is about 20 times larger than that of D atom. The data were taken by AGNES spectrometer of ISSP (installed at JRR-3, Tokai). Broadening of elastic peak due to quasielastic scattering appeared in the temperature range above 280 K. The data were fitted well by two Lorentz functions as shown in Fig. 2; the result of fitting is given by a black curve and a broad component by a blue curve. The sharp (slow) component corresponds to ionic diffusion while the broad (fast) one maybe intraionic motions. By analyzing the Q dependence of the quasielastic data, it was found that the ionic diffusion in bulk EMITFSI is represented by a simple Brownian motion while that in the ion gels by the jump-diffusion model. We are now preparing the neutron scattering experiments with a high resolution spectrometer IRIS in Rutherford-Appleton laboratory (UK) to investigate the motion of PMMA which is much slower than that of EMITFSI.

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Spin Correlations and Excitations in the Transition-Metal-Based Quasicrystal Zn-Fe-Sc

Sato Group

Quasicrystal is a yet mysterious form of solids, differingfrom periodic crystals and random glasses. Structure of the quasicrystals is now understood using an idea of quasiperiodicity, which has hidden translational order in higher dimensional space. Magnets with quasiperiodic structure are of particular interest because they may exhibit unique collective behavior that cannot be seen in the periodic crystalline systems. A fundamental issue of quasicrystal magnetism is formation of localized moments, and interplay of the localized moments and conduction electrons, in the 3*d*-transition-metal-based systems. The recently discovered Zn-Fe-Sc icosahedral quasicrystal provides an opportunity to study this issue [1].

Magnetic susceptibility of the Zn-Fe-Sc quasicrystal follows the Curie-Weiss (CW) law in the wide temperature range 76 < T < 300 K. An effective moment of Fe estimated from the CW fit is extraordinary large as $\mu_{\rm eff} = 5.3 \,\mu_{\rm B}$. This suggests that the Zn-Fe-Sc quasicrystal is the first magnetic quasicrystal with stable 3*d* magnetic moments. Below $T_{\rm f} = 7.2$ K, an irreversibility is seen for the field-cooled and zero-field-cooled magnetizations. Hence, the spin freezing may likely take place at $T_{\rm f}$, as commonly seen in the magnetic quasicrystals.

Compared to the 4f electrons of the rare-earth elements,

Fig. 1. Neutron diffraction patterns at T=200K and 4K. Difference between the data at the two temperatures is also shown.

wave functions of the 3d electrons have larger spatial extent with energies closer to the Fermi level. This may possibly result in different single-site spin fluctuations as well as intersite spin correlations in the Zn-Fe-Sc quasicrystal from those in the rare-earth-based quasicrystals. Hence, to reveal intrinsic spin correlations and excitations in the 3d transition-metal-based system, we performed neutron scattering investigation on the Zn-Fe-Sc quasicrystal [2].

Figure 1 shows the neutron diffraction patterns measured at the paramagnetic temperature T = 200 K and the frozen temperature 4 K using the triple-axis spectrometer ISSP-GPTAS in the triple-axis mode. At the high temperature, only the nuclear Bragg reflections were observed; the peak positions are consistent with the quasicrystalline structure. On the other hand, at the frozen temperature, weak increase of the elastic intensity was observed, indicating development of the short-range spin correlations. The temperature difference, shown in the figure, much clearly detect the development of the short-range correlations; a broad peak around Q = 0.6Å⁻¹ can be seen.

The inelastic spectra were measured at several temperatures at Q~0.72Å⁻¹ to obtain insight into the 3*d* spin dynamics. Figure 2 shows the resulting inelastic spectra. At the paramagnetic temperatures the broad quasielastic scattering was observed in addition to the huge elastic signal. As temperature is decreased, the quasielastic signal becomes narrower and weaker. Nevertheless, prominently the quasielastic signal survives below the macroscopic spin

Fig. 2. Inelastic scattering spectra at several temperatures between 100K and 4K.

freezing temperature $T_{\rm f}$. We note that the energy scale of the remaining spin fluctuations (estimated from the width of the quasielastic signal) is larger than the thermal energy $k_{\rm B}T$. Therefore, the origin of the remaining spin fluctuations may be quantum mechanical; interaction between the conduction and 3d electrons may be the key for understanding the remaining base temperature fluctuations.

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Quantum Phase Transition in Spin-1 Chain System SrNi₂V₂O₈ Induced by Cation Substitution

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Quasi-one-dimensional (1D) spin-chain systems have been actively studied in condensed matter physics. In general, an ideal 1D spin-chain system does not show long-range ordering (LRO) above T = 0 K due to strong quantum spin fluctuation, however almost all quasi-1D compounds display LRO governed by weak interchain interactions (J_{\perp}). Haldane [1] predicted that a spin-1 Heisenberg antiferromagnetic chain system displays a spin-liquid state with an energy gap (Haldane gap), while an uniform spin-1/2 chain system does a spin-liquid state but a gapless continuum of excitations. Sakai and Takahashi [2] further proposed a theoretical phase diagram in the D- J_{\perp} plane for

Fig. 1. Crystal structure of tetragonal $ANi_2V_2O_8$ (A = Sr and Pb) viewed along the *c*-axis (a) and the *a*-axis (b). The screw chains formed by edge-shared NiO₆ octahedra (blue) are separated by VO₄ tetrahedra (gray) and Pb²⁺/Sr²⁺ ions (black). (c) Sakai-Takahashi phase diagram for the ground state of 1D spin-1 chain systems.

Fig. 2. Magnetic susceptibility and heat capacity of pure $SrNi_2V_2O_8$ (black), $Sr_{0.95}Ca_{0.05}Ni_2V_2O_8$ (brown) and $Sr_{0.7}Ba_{0.3}Ni_2V_2O_8$ (blue). The ground state of $SrNi_2V_2O_8$ is a spin liquid state (magnetically disordered state). A minor substitution of Ca^{2+} with smaller ionic radius for Sr^{2+} changes the ground state of $SrNi_2V_2O_8$ to a Néel-like ordered state. On the other hand, Ba^{2+} ion-substitution stabilizes spin-liquid state much more.

the ground state of 1D spin-1 chain systems and suggested that a quantum phase transition between a spin-liquid state and Néel-like ordered state occurs as the interchain interaction J_{\perp} and/or single ion anisotropy *D* reach beyond a certain critical value.

Recently, 1D spin-chain system PbNi₂V₂O₈ and SrNi₂V₂O₈ have attracted much attention in this respect. A significant structural feature is that all magnetic Ni²⁺ ions (S = 1) are equivalent with the arrays of edge-shared NiO₆ octahedra forming screw chains along the c-axis, and the screw chains are separated by nonmagnetic V5+O4 tetrahedral and Pb²⁺/Sr²⁺ ions, resulting in a quasi-1D structural arrangement (Fig. 1(a) and (b)). Uchiyama et al. [3] reported that PbNi₂V₂O₈ is a Haldane-gap system, while SrNi₂V₂O₈ shows a three-dimensional (3D) Néel ordered state with weak ferromagnetism below 7 K. Zheludev et al. [4] suggested, from inelastic neutron scattering studies, that PbNi₂V₂O₈ and SrNi₂V₂O₈ are close to the phase boundary on the Sakai-Takahashi phase diagram in which PbNi₂V₂O₈ is located on one side with a spin-liquid ground state, whereas SrNi₂V₂O₈ is likely on the other side with 3D Isinglike Néel ordered state (Fig. 1(c)). However, on the basis of an NMR study, Pahari et al. [5] recently suggested that SrNi₂V₂O₈ is likely a Haldane spin system with a nonmagnetic ground state similar to that seen in PbNi₂V₂O₈.

The different conclusions have led us to investigate quantum critical behaviors of SrNi₂V₂O₈ by a minor substitution of Ba^{2+} with a larger ionic radius or Ca^{2+} with a smaller one for Sr^{2+} [6]. We found no λ -like feature in the heat capacity data of pure SrNi₂V₂O₈ from 300 to 1.9 K, suggesting that SrNi₂V₂O₈ falls likely into a spin-liquid ground state (Fig. 2). Also, a Néel ordered state was clearly observed in $Sr_{0.95}Ca_{0.05}Ni_2V_2O_8$ with 5% Ca^{2+} ions-substitution in the system (Fig. 2). The magnetic susceptibility data support these results (Fig. 2). A slight change of only ~0.01Å in the lattice constant a between SrNi₂V₂O₈ and Sr_{0.95}Ca_{0.05}Ni₂V₂O₈ induces a quite different ground state, providing concrete evidence that SrNi₂V₂O₈ is indeed close to the phase boundary between a spin-liquid and Ising-like ordered states on the Sakai-Takahashi phase diagram. We also observed that Ba2+ ion-substitution stabilizes spin-liquid

state much more (Fig. 2). In conclusion, $SrNi_2V_2O_8$ locates in the spin-liquid ground state side that is close to the phase boundary on the Sakai-Takahashi phase diagram and a quantum phase transition of a spin-liquid state to an ordered state is induced by a minor substitution of Ca^{2+} ions.

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What Happens in Electronic State of β -A_{0.33}V₂O₅ around Critical Pressure?

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Electronic pressure (*P*)-temperature (*T*) phase diagrams of a series of quasi-1-dimensional conductors, β -vanadium bronzes β -A_{0.33}V₂O₅ (A = Li, Na and Ag), are particularly interest. As shown in Fig. 1(a), charge ordering (CO) phases are suppressed by high pressure and superconducting (SC) phases appear in the vicinity of CO phases. In order to gain a deeper insight into the pressure-induced superconductivity of β -A_{0.33}V₂O₅, the resistivity (ρ) under high pressure has been carefully measured on the precisely tuned stoichiometric samples and the obtained data have been analyzed. Figure 1(b) shows the ρ versus T^2 curves for all β -A_{0.33}V₂O₅ near the critical pressure (P_c) and up to 70 K ($T^2 = 4900$ K²). As clearly seen in Fig. 1(b), normal metallic (NM) phases show $\rho = \rho_0 + AT^2$ behaviors around P_c region, indicating their Fermi liquid-like nature.

The pressure dependences of two fitting parameters Aand ρ_0 are exhibited in Fig. 1(c). The linear fitting for each compound was carried out in the restricted pressure region where linear relation of ρ - T^2 curves was realized. The white, light gray and dark gray backgrounds in this panel represent the regions with CO, CO+SC, and SC ground states, respectively. The white region in higher pressure side for Li-compound corresponds to another non-SC higher pressure phase (NP phase) where both SC and T^2 behavior disappear. The most significant feature is that the pressure dependence of coefficient A (A-P curve) seems to have a peak at around $P_{\rm c}$. Meanwhile, the residual resistivity ρ_0 monotonically decreases with increasing pressure. A clear peak of A-P curve was observed only in Na-compound. We cannot know the behavior in the higher pressure side than P_c for Li-compound because it is probably masked by NP phase. On the other hand, the region of T^2 dependence below $P_{\rm c}$ is too narrow to evaluate the behavior for Ag-compound. However such peak behavior of A-P curve is naturally expected in both Li- and Ag-compounds on the analogy of Na-compound.

In the ordinary Fermi liquid picture, the coefficient *A* and residual resistivity ρ_0 are proportional to $(m^*)^2$ and $1/lS_F$, respectively, where the m^* , *l* and S_F are effective mass, mean free path and area of Fermi surface, respec-

Fig. 1. (a) Electronic *P-T* phase diagrams of β -A_{0.33}V₂O₅ (A = Li, Na and Ag). NM, CO, and SC denote *normal metallic, charge ordering,* and *superconducting* phases, respectively. NP in Li-compound means another non-SC higher pressure phase in which both SC and T^2 behavior of resistivity (ρ) disappear. (b) ρ versus T^2 plots for all compounds near the critical pressure P_c up to 70 K. The $\rho = \rho_0 + AT^2$ behaviors in NM phases are observed in relatively wide pressure dependences of residual resistivity ρ_0 (open symbols) and *A* coefficient of T^2 term (closed symbols). The white, light and dark gray backgrounds refer to the region of CO phase, coexistence of Li-compound in higher pressure side corresponds to NP phase.

tively. Thus, A and/or ρ_0 are naturally expected to be relevant to superconducting temperature $T_{\rm SC}$. The relation $\rho_0 \propto 1/lS_{\rm F}$ suggests considerable increase of the $S_{\rm F}$ by a factor five around $P_{\rm c}$ (*l* is naturally expected to be constant with increasing pressure). On the other hand, the peak behavior of *A-P* curve demonstrates an enhancement of effective mass $(m^*)^2$ by a factor two (Na-compound) at $P_{\rm c}$. Such enhancement of the coefficient *A* is probably attributed to some increase of the interaction between carriers via charge fluctuation near $P_{\rm c}$.

The most noteworthy aspect is that the superconducting transition temperature $T_{\rm SC}$ decreases with increasing pressure even though ρ_0 decreases, namely, $S_{\rm F}$ probably increases with pressure. This strongly implies non BCS-type superconductivity. Moreover, $T_{\rm SC}$ as a function of pressure seems to scale to $A \propto (m^*)^2$, indicating that the correlation between electrons is an important parameter for $T_{\rm SC}$.

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A New Quantum Monte Carlo Method and Laser Trapped Bosoninc Atoms

Kawashima Group

The phase transition from a superfluid to a Mott insulator in bosonic systems has been intensively investigated since it was observed in an ultra cold Bose gas trapped on an optical lattice. [1] The atomic gas was trapped in a harmonic potential and a periodic lattice potential generated by laser beams. This system can be well described by Bose-Hubbard model (BHM)

$$H = -\frac{t}{Z} \sum_{\langle i,j \rangle} \left(b_i^{\dagger} b_j + b_j^{\dagger} b_i \right) + \frac{U}{2} \sum_i b_i^{\dagger} b_i^{\dagger} b_i b_i - \mu \sum_i b_i^{\dagger} b_i,$$

where b_i (b_i^{\dagger}) is the boson destruction (creation) operator at a site *i* and Z=6 is the coordination number in the cubic lattice. In the optical lattice system, the ratio of the transfer integral *t* and the repulsive interaction *U* can be controlled simply by tuning the intensity of the laser beams, which makes this system an ideal laboratory for investigating quantum many-body problems. In order to first discuss the thermodynamic properties, we neglect the gradient of the chemical potential corresponding to the trapping potential. We also choose $\mu/U=1/2$ in order to study a typical case of the phase transition.

Here, we present the results of the quantum Monte Carlo (QMC) simulation of BHM using a modified directedloop algorithm [2]. The directed-loop algorithm is one of the most widely applicable algorithms for QMC based on the Feynman path-integral. The modification makes the algorithm very efficient in particular for the bosonic systems. We show the finite temperature phase diagram in Fig. 1. For large t/U at fixed temperature, the system is in the superfluid phase. When t/U is decreased and reaches a certain value, a phase transition to a normal gas phase occurs. As a function of the transfer integral, the transition temperature decreases as t/U decreases, and eventually it vanishes at the quantum critical point. Beyond this point, the system at T=0 is in the Mott insurating phase, with an excitation gap increasing with decreasing t/U.

In the time-of-flight experiment, the column-integrated momentum distribution is observed as an interference

Fig. 1. The superfluid transition temperature T_c (red) and the singleparticle energy gap Δ in the Mott insulator (blue) at $\mu/U=1/2$. They both vanish at the quantum critical point t/U=0.192(2). The typical interference patterns are also shown. The corresponding lattice depth V_0 in units of the recoil energy E_R is indicated on the top axis. $(E_R=h^2/8Md^2$, where M is the mass of the rubidium atom and d is the lattice constant.) [3]

Fig. 2. Column-integrated momentum distribution $N_{\perp}(k_x, k_y)$ and its $k_y=0$ profile $N_{\perp}(k_x, k_y=0)$ at a temperature slightly above T_c . The parameters are t/U=0.25, $\mu/U=1/2$, $T_c/t=0.7$. The lattice size is 12³. [3]

pattern. The corresponding quantity in the QMC is

$$N_{\perp}(k_x,k_y) = \int dk_z |W(\mathbf{k})|^2 n(\mathbf{k})$$

where $W(\mathbf{k})$ is the Fourier transformed Wannier function and $n(\mathbf{k}) = \sum_{i,j} \langle b_i^{\dagger} b_j \rangle \exp\{i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)\}$. In Fig. 1, three typical cases, *i.e.*, the superfluid, Mott insulator, and critical, are shown. As is naturally expected, we see sharp peaks in the pattern in the superfluid region, while they do not appear in the Mott insulator region. The sharp peaks, therefore, have been regarded as a clear indicator of the superfluidity, and their disappearance have been thought to occur at the quantum phase transition to the Mott insulator phase. However, in Fig. 2 we see rather sharply peaked interference pattern near the critical temperature (t/U=0.25 and $T=1.1T_c$) but still clearly in the normal gas phase. In this region, the superfluid density is zero. Nevertheless, the sharp peaks are present. The width of the sharp peaks in the normal region corresponds to the inverse of the correlation length. Near the critical temperature, the correlation length is large and the peaks, therefore, tend to be so sharp that it cannot be distinguished from delta-function peaks that truely signify the superfluidity. Thus, our results show that the interference pattern with sharp peaks is not necessarily a reliable indicator of superfluidity.

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Renovation of World Record of In-door Highest Magnetic Field, 670 Tesla, and Its Application to Solid State Materials

Takeyama Group

We continued reconstruction of our electromagnetic flux-compression (EMFC) system including the coil structures, and we are renewing records of the highest magnetic field successively form 630 Tesla, 650 Tesla, and recently 670 Tesla, by 4 mega joule injection out of a 5 mega joule condenser bank. All these records of magnetic field are presented in Fig.1. Our recent progress of the records is characterized by the reproducibility and reliability of the data in comparison to the previously addressed data, which were somehow obtained accidentally.

Using the new EMFC system, we started measurements of solid state materials. One of the subjects is the observation of full-magnetization processes of the chromium spinel, ZnCr₂O₄, which poses interesting magnetic order arising from the intense geometrically frustrated antiferromagnetic spins at pyrochlore lattice sites. According to the existing theories [1], magnetic fields more than 300 Tesla are necessary to achieve the full saturation of the magnetization in this material with such a high Curie-Weiss temperature, -390 K but with a fairly low Neel temperature, 12.5 K. The rotation angle showed a well-resolved signal linearly proportional to a magnetization with the cant 2:1:1 magnetic phase occurred around 120 Tesla as well as the 1/2 plateau between 130-160 Tesla as shown in Fig.2. The cant 2:1:1 phase is theoretically predicted to appear for the system with relatively weak spin lattice interaction. For a precise Fadaray rotation (FR) measurements in the EMFC system, we have developed a hand made optical cryostat fully made of a "stycast" resin, which enables to achieve low temperatures with a fairly good vacuum tight. The measurement precision was improved also by fixing a vacuum chamber, in which the copper liner and the optical liquid He-flow cryostat are inserted, directly to the primary coil (Inset in Fig.2). Figure 2 showed one of the successful FR data measured up to 90 % of the full saturation measured up to 350 Tesla at 17 K, using the EMFC system. The quality of the data is comparable to those obtained by the single turn coil, and also even with those obtained by the induction method in the non-destructive long pulse magnet previously performed [2].

Fig. 1. Magnetic field traces of recent EMFC experiments. The Cu lining primary coil, which has been newly designed and improved the energy transfer efficiency, is shown in the backdrop.

Fig. 2. Magnetization of $ZnCr_2O_4$ measured by the Faraday rotation method. Red colors are measured in the single turn coil system, and blue one in the EMFC system. The dotted line is calculated by K. Penc [1]. The region of the respective magnetic phase is indicated. The inset shows an experimental setup around the primary coil with an optical cryostat.

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Authors

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Magnetization Process and the Associated Lattice Deformations in an Intermetallic Compound Gd₅Ge₃

Kindo Group

The interplay between magnetism and lattice degrees of freedom in solids sometimes produces novel phenomena. Gadolinium germanides and silicides have attracted considerable interest because of their potential application to magnetic cooling [1-4]. In particular, $Gd_5(Si_2Ge_2)$ shows a giant magnetocaloric effect [1]. The compound, Gd_5Ge_4 exhibits a massive magnetic-field-induced structural transformation which is an origin of the giant magnetocaloric effect [3]. We have been developing an X-ray diffraction technique under a pulsed magnetic field [5] which gives a unique opportunity to explore new phenomena in which a lattice degree of freedom plays an important role in high field region. We study the nature of the coupling between magnetism and lattice in a related compound Gd_5Ge_3 . The material Gd_5Ge_3 has the hexagonal crystal structure

with space group $P6_3/mcm$. Gd atoms occupy the crystallographic sites, 4(d) and 6(g) [6]. These Gd atoms form repeated layers along the *c* axis. The magnetic susceptibility, χ , at high temperatures obeys the Curie-Weiss law with Weiss temperature, Θ , = 54.8 K and the χ of a single crystal of Gd₅Ge₃ exhibits two anomalies at $T_{\rm N}$ = 76 K and $T_{\rm t}$ = 52 K [7]. In this report, the results of magnetization and synchrotron X-ray diffraction measurements on Gd₅Ge₃ under pulsed high magnetic fields are reviewed.

A single crystal of Gd₅Ge₃ was grown by the Czochralski method at Hiroshima University. The magnetization, M, up to 7 T was measured with a Quantum Design's MPMS SQUID magnetometer and M up to 58 T was measured with a pulsed field magnet at ISSP, the University of Tokyo. Synchrotron X-ray diffraction measurements in pulsed high magnetic fields were performed at the beamline BL19LXU of SPring-8. For magnetic field, H, applied along the aaxis, M increases linearly with H up to around 2.2 T and exhibits a small jump. Then, M shows a steep increase at $H_{\rm C}$ = 3.8 T. The magnetization change at $H_{\rm C}$ is seen in both of the increasing and decreasing fields, but that at about 2.2 T is not seen in the decreasing field. Along the c axis, M increases monotonically with H after exhibiting a small jump around 2.8 T. This change in M is not also seen in a decreasing field. The main panel of Figure 1 shows the

Fig. 1. The magnetic field dependence of the lattice constant (a) a and (b) c of Gd₅Ge₃ measured at T = 4.2 K. The external magnetic field *H* is applied along the a axis. Curves are drawn as a guide to the eye.

Inset: (a) The magnetic field dependence of the magnetization when H is applied along the a axis. (b) The crystal structure and a model magnetic structure of Gd₅Ge₃. Short arrow shows the individual spin of Gd atom and large arrow represents the trimer spin.

magnetic field dependence of the lattice constants determined from the (5 5 0) and (0 0 8) reflections. The lattice constant a elongates with increasing H. In contrast, the lattice constant c shrinks with increasing H.

Here, we propose a model magnetic structure in which the three nearest-neighboring Gd spins on the 6(q) site form a ferromagnetic trimer and the trimers form an antiferromagnetic triangular lattice (AFTL) in the a-a plane. Since Gd^{3+} spin (S = 7/2) is regarded as a classical one, we expect that a 120° spin structure will be realized in the *a*-*a* plane in zero field. In this magnetic structure, the exchange field acting on a Gd spin on the 4(d) site from the neighboring spins on the 6(q) site is zero. Then, Gd spins on the 4(d)site order antiferromagnetically in the a-a plane forming a honeycomb lattice, independent of the AFTL. Both magnetic moments on the 6(g) and 4(d) sites are coupled ferromagnetically along the c axis. We interpret that the transition from the 120° to ferrimagnetic phase and that from the ferrimagnetic to canted phase occur at H_C simultaneously, because the change of the magnetization is about 1.5 μ_B which is nearly equal to one-third of the saturation magnetization of Gd spins on 6(q) site. A calculation based on the proposed magnetic structure and possible exchange paths explains successfully the opposing behavior of the lattice deformations and the M^2 dependence. The essential ingredient to explain the opposing behavior of the lattice deformations is the presence of the antiferromagnetic (in the a-a plane) and ferromagnetic (along the c axis) exchange interactions.

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Novel Multiferroic States in Eu1-xYxMnO3

Tokunaga Group

Recent discovery of gigantic magnetoelectric effects in perovskite-type RMnO₃ (R = Tb and Dy) shed a new light on the physics in ferroelectric magnets [1,2]. In this class of materials, non-collinear spin ordering in the frustrated Mn-spin system is believed to be the origin of the ferroelectricity. The effect of magnetic fields (*H*) on this frustrated system is, however, difficult to be figured out because presence of 4*f* moments in the rare-earth ions complicates the system. This difficulty can be removed with using a mixture of non-magnetic Eu³⁺ and Y³⁺ for the R-site. Actually Eu_{1-x}Y_xMnO₃ realizes multiferroic states when average ionic radius of the R-site becomes comparable to that of Tb [3]. Recent X-ray diffraction measurements revealed that application of *H* flops the cycloidal

Fig. 1. Magnetic field (H) dependence of electric polarization (P) in EuMnO₃ at various temperatures. Application of H|b higher than 20 T induces P||a. Further increase in $\hat{H}||b$ causes discontinuous changes in P accompanied with hystereses. These transitions coincide with metamagnetic transitions (not shown).

spin structure, and hence electric polarization (P) coupled with it, which can be intuitively understood as a reduction in the Zeeman energy [4]. It is, however, not clear whether this magnetoelectric effect in low-field alone represents the observed variety of field-effects in RMnO₃.

To study more general nature of magnetic field-effects on this frustrated-spin system, we studied magnetic and dielectric properties in single crystals of Eu1-xYxMnO3 $(0 \le x \le 0.4)$ using pulsed-magnetic fields up to 55 T. The experimental results on EuMnO₃ demonstrate the presence of first-order transitions from paraelectric to ferroelectric P||a phase when H higher than 20 T is applied along the b-axis (Fig. 1). Similar field-induced transitions are also observed in a sample with x = 0.4 at various temperatures (T), in which spontaneous P exists along the a-axis even in zero magnetic field. This fact indicates that high-field P||a|phase is different from the known *ab*-cycloidal state in low fields [3,4]. Judging from the similarity in the P-H curves and in the stability region in the H-T plane, we consider the high-field P||a phases in x = 0 and 0.4 as an identical electromagnetic phase.

Now we discuss the origin of the field-induced P||a|phase. On the analogy of the field-effects in the low-field region, we assume the spin-flop phenomenon as the origin of this transition. According to the microscopic theoretical model that can reproduce multiferroic states in RMnO₃ in

Fig. 2. Magneto-electric phase diagram of $Eu_{1-x}Y_xMnO_3$ in the H-T plane for several values of x. Novel P||a phase (orange area) extends over high-field region of canted-antiferromagnetic, sinusoidal, and ab- and bc-cycloindal phases.

Fig. 3. Relationship between P and M in Eu_{0.6}Y_{0.4}MnO₃ derived from the M-H and P-H curves at 4.2 K. Quadratic relation between P and M is observed in the ab-cycloidal phase (left-panel), but not in the novel phase ($M \ge 1 \mu_B/Mn$ region in the right panel).

low-fields, P changes as a function of M as $P \propto 1 - (M/M_s)^2$ when the cycloidal-cone closes by applied H (see insets of Figs. 3), where M_s denotes the saturation moment. This relation is observed in the low-field P || a phase in H || c, while not in the high-field P||a phase in H||b (Fig. 3). This discrepancy suggests that the novel P||a phase in high-fields has different microscopic origin from that in low-fields.

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Highly Efficient and Brilliant High Harmonic Coherent Soft X-Ray Laser Source from Laser-Ablated Solid **Target Plasma towards** a Water Window Region

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 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

Fig. 1. HH spectra from tellurium and silver laser-ablation plumes were obtained at the wavelength of 20 nm. A strong 27th harmonic at the wavelength of 29.44 nm was obtained.

spectroscopy and ultrafast dynamics of multiphoton-induced photoelectron emission has recently been reported. Thus the High-order Harmonics 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 shorter wavelength region (high photon energy region).

For the high intensity HH, we report first demonstration of the single harmonic enhancement by using by multiphoton resonance with the strong oscillator strength ions at the wavelength of 47 and 37 nm. Furthermore we have demonstrated the intensity enhancement of single harmonic at 29.44 nm using the laser-ablation tellurium plume [1]. The enhancement of a single harmonic is due to multiphoton resonance with a Te II transition having strong oscillator strength. By tuning the wavelength of the pump laser, we found that the enhancement factor depends on

Fig. 2. HH spectra obtained from the gold plasma at different chirps and pulse durations of the driving radiation. The line connecting the 49th harmonic at different chirps shows a blue shift in the case of chirp-free pulses. The signs of pulse duration values indicate the sign of the chirp.

the difference between the wavelength of the enhanced harmonic and that of the strong radiative transition. For tellurium, we show that intense single harmonic generation can be generated when the wavelength of the 27th harmonic is closer to the wavelength of the strong transition. 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

Various plasmas prepared by laser ablation of the surfaces of solid targets were examined by the narrowband width radiation of different chirp and pulse durations. Figure 2 shows the HH spectra from the gold plasma at different chirps. The high-order harmonics generated during laser plasma interaction showed different brightness, wavelength shift, harmonic cutoff, and efficiency by using variable chirps of pump radiation. An analysis of harmonic optimization at these conditions is presented. The blue shifted and red shifted harmonics observed in this case were analyzed and attributed to the abundance of free electrons and self-phase modulation of the driving pulse. The resonance-induced enhancement of the 15th harmonic from GaN-nanoparticle-containing plasma caused by the tuning of harmonic wavelength close to the ionic transition was demonstrated.

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Fabrication of Two-Dimensional Periodic Nanostructures by Two-Beam Interference of Femtosecond 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 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.

Two-dimensional periodic nanostructures on ZnO crystal surface were fabricated by two-beam interference of 790 nm femtosecond laser. The long period is, as usually reported, determined by the interference pattern of two laser beams. Surprisingly, there is another short periodic nanostructures with periods of 220-270 nm embedding in the long periodic structures. We studied the periods, orientation, and the evolution of the short periodic. 2D periodic nanostructures were fabricated in ZnO crystal surface induced by two-beam interference of fs laser beam [1].

Fig. 1. SEM images of 2D periodic nanostructures. The total pulse energy and irradiation time were 127 μ J and 6 s in (a -c), 154 μ J and 4s in (d), and 250 μ J and 10 s in (e), respectively.

Figures 1(b) and 1(c) show the SEM images in the white square marked in Figs. 1(a) and 1(b) at higher magnifications, respectively. These photos surprisingly represented 2D periodic structures. As reported update, only one dimensional periodic structure was obtained by two-beam interference method, and the period was determined by the interference pattern: $\Lambda = \lambda / 2\sin\theta$. The laser wavelength λ is 790 nm, and the angle between the two laser beams is $2\theta = 35.2^{\circ}$. The period is estimated to be 1.31 μ m. The long period is 1.33 μ m (see Figs. 1(b) and 1(c)), which is in accordance with the theoretical value. Therefore, the long periodic structures are determined by the interference pattern of the two laser beams.

Besides the long periodic structures, there are short periodic nanostructures (nanogratings) embedding in the long ones. The nanograting is of 1.14 μ m wide, and its period is only 250 nm. If the total pulse energy increased to 154 μ J and the irradiation time decreased to 4 s, the nanograting width decreased to 1 μ m, and its period increased to 270 nm (shown in Fig. 1(d)). We adjusted the laser conditions, and found the width of the short periodic structure changed in the range of 0.4-1.15 μ m while the long period kept as 1.33 μ m. If the pulse energy was higher than 240 µJ, the nanogratings were ablated. However, on the interval line between each nanogratings, periodic nanospots of 0.5 μ m long and 0.25 μ m wide appeared (Fig. 1(e)). In one square centimeter in the sample surface, there was about $(1/1.33) \times (1/0.25) \times 108=3 \times 108$ cm⁻² nanoripples or nanospots, and the short period was only 250 nm. Therefore, the 2D periodic nanostructures have great potential applications in ultra-high density optical record and photonic crystals in ultraviolet and visible light range.

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 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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