Summary of A01 Planned Research Group -Correlation effect between localized multipoles and conduction electrons-

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The subject of A01 planned research group is to study the novel quantum phenomena driven by the strong hybridization between localized mulitipoles and conduction electrons. I will review the recent activities made by A01 group during the four and half year period. Our study covers not only the atomic multipoles but also the cluster multipoles. The materials that have been covered by our project is not only *f*-based heavy fermions but also *d*-electron systems. The talk will cover the topics related to (1) multi-channel Kondo effect due to multipoles and exotic superconductivity, (2) singular charge fluctuations near quantum criticality, (3) valence quantum phase transition, (4) anomalous Hall effects in antiferromagnets, (5) anomalous Hall effect due to cluster multipoles, (6) f-electron based skyrmion, (7) novel cluster multipole order (8) quasi-crystals superconductivity.

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Summary from B01 group

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The target of B01 group is to explore the various conducting phenomena and quantum phases induced by the "itinerant multipole". The main target materials are 5f-electron systems which has the dual nature between itinerancy and localization. The 5f electron has the multiple degrees of freedom for multipoles due to the strong spin-orbit coupling with the parity mixing with 6d electrons. We summarize our results during J-Physics project, which include ferromagnetic superconductivity URhGe, UCoGe, UGe_2, the novel spin-triplet superconductor UTe_2, UBe_{13}, URu_2Si_2, topological phenomena, Bi chalcogenides, UNi_4B, and so on. Some of the results were obtained through the collaborative research between different project groups, and the international collaborations.

Summary of C01 Planned Research Group

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The Planned-Research Group C01 has been conducting experimental and theoretical studies on static and dynamical properties of *augmented multipoles*, which have been introduced and developed in the J-Physics project as extension of *traditional multipoles* formed by a single orbit on a single ion. One of the most important advances made in our study would be the theoretical formularization of the concept of the augmented multipoles, where they are clearly defined, consisting of two types of extension: *cluster multipoles* formed from spin and charge distribution on plural atomic sites and *hybrid multipoles* formed from multi-orbits around an ion [1]. On this basis, extensive theoretical analyses and predictions have been made on a variety of actual materials containing not only *f* electrons but also *d* and *p* electrons [2]. The investigation of a variety of cross correlations among magnetic, electric and elastic degrees or freedom driven by emergence of the cluster multipoles have now been intensively conducted not only in C01 but also in other groups. As materials investigated in this line so far, UNi_4B [3-5], α-$Cu_2V_2O_7$ [6,7], $Co_4Nb_2O_9$ [8,9], $(LaMnO_3)_2(SrMnO_3)_2$ [10], $BaMn_2As_2$ [11], Mn_3Sn [12], $ErNi_3Ga_9$ [13], $TbNiC_2$, StPtAs [14] are exemplified. In StPtAs, particularly, a possibility of the chiral *d*-wave superconductivity has carefully been examined [15]. We have also contributed the development of understanding of unusual properties in chiral compounds, such as skyrmion, chiral soliton, and non-reciprocal responses, through resonant X-ray scattering measurements [16, 17] and material development [18]. Regarding the development of experimental techniques, we have developed the precise magnetization measurements under electric currents have been developed, and suggested that this technique can be a useful tool to detect a hidden broken space-inversion symmetry of a system. Moreover, very recently, it has been found that X-ray magnetic circular dichroism (XMCD) can be an effective approach for analyzing augmented multipoles in antiferromagnets [19]. We should further expand and deepen the studies of this trend, exploring new functions in view of new application. It will also be desired to make a careful search for the substances where hybrid multipoles emerge as hidden order parameters. It will be very important to establish a typical example of such systems, demonstrating the direct observation of hybrid orbitals as well as precise structural refinement. Significant progress should be made on the development of new materials that provide a playground with augmented multipoles.

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Spin textures of Conduction Electrons in Skyrmion Compound EuPtSi

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EuPtSi is a skyrmion compound with a same space group as in MnSi. Fundamental magnetic properties are almost the same between two compounds [1-3]. The magnitude of the propagation vector $\boldsymbol{q}$ = (0.2 0.3 0) is, however, large and then a size of the skyrmion $\xi = 2\pi / |\boldsymbol{q}| = 18$ Å is one order smaller than 180 Å in MnSi. The skyrmion phase is therefore not found in any field direction. We succeeded in growing high-quality single- crystals of EuPtSi and have measured the de Haas- van Alphen (dHvA) oscillations.

Figure 1(a) shows the angular dependences of dHvA frequencies *F*. *F* corresponds to the maximum or minimum cross-sectional area of the Fermi surface. The largest Fermi surfaces, which are named α and α', are spin-split reflecting the noncentrosymmetric (cubic chiral) structure. The Fermi surfaces are not spherical but cubic in shape. Figures 1(b)-1(e) correspond to (001) and (111) cross-sections and the corresponding directions of spins for the conduction electrons are illustrated.

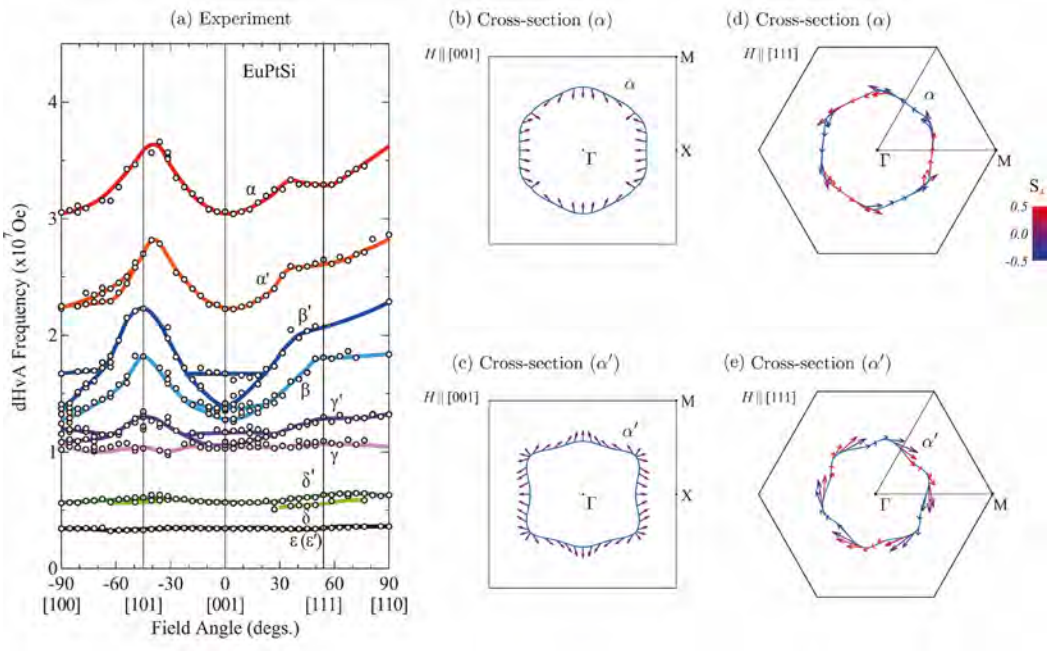

Fig. 1. (a) Angular dependences of dHvA frequencies in EuPtSi, (b) and (c) spin textures in the cross-sections in the (001) plane, and (d) and (e) in the (111) plane in SrPtSi.

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EuPtSi: thermodynamic properties

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There is growing interest in the chiral helimagnet EuPtSi, since a skyrmion lattice phase (A phase) appears in the intermediate field range in the ordered state below T_N=4.0 K [1-3]. We studied the phase transitions in EuPtSi by means of high-precision magnetization measurements.

The helimagnetic transition in EuPtSi has been reported to be weakly first order [1,2,4] at low fields. We examined the field evolution of the helimagnetic transition by magnetization measurements [5]. At low fields, the temperature derivative of the magnetization dM/dT exhibits a sharp symmetric peak at T_N (H), indicating a first-order transition. By increasing H, the shape of dM/dT drastically changes above a certain field to an asymmetric step-like function. The results provide strong evidence for a tricritical point, whose location is found to be strongly field-orientation dependent. Field variation of M in the paramagnetic state above T_N indicates that strong antiferromagnetic fluctuations exist and persist up to ~7 K, suggesting that the first-order transition is driven by strong fluctuations of the order parameters [4-6]. This situation, in which the single-$\boldsymbol{q}$ ordering temperature is suppressed much below a mean-field transition temperature, is favourable for the occurrence of multi-$\boldsymbol{q}$ orderings like the skyrmion phase.

The A-phase is characterized by a quasi-plateau in the field variation of M. For H || [111], the A-phase is entered from the conical phase accompanied by a sharp peak in dM/dH; the phase transition is of first order. The A-phase is field induced at finite temperatures above ~0.4 K. In the ground state below 0.3 K, the A-phase does not appear in a zero-field-cooled condition, and the conical-state magnetization linearly increases with H and saturates near 3 T for all directions. When field-cooled at 1.5 T from well above 0.3 K in the A-phase, however, the A-phase persists down to the base temperature of 50 mK. The results imply that the low-temperature boundary of the A-phase is strongly first order, and the A-phase can survive as $T \rightarrow 0$ in a supercooled condition. The magnetization for H || [100] also shows very similar behavior.

No A-phase, however, appears for H near the [110] direction. This point might be related to the fact that [110] is nearly parallel to the helical propagation vector, $\boldsymbol{q}$=(0.2,0.3,0). In general, the helical state is most stable when $H \parallel \boldsymbol{q}$. If $\boldsymbol{q}$ is pinned to the lattice [3], then the helical state might prevail over the A phase in H near the [110] direction. This point is consistent with the short wavelength of the order parameter of this system is relatively short, since the Fourier-transformed exchange energy generally becomes anisotropic in k space when k is large.

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Resonant X-ray Scattering of chiral antiferromagnet EuPtSi

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EuPtSi has a chiral crystal structure belonging to the space group $P2_13$ (no. 198, T^4) that has no inversion center, and causes magnetic ordering due to divalent Eu at $T_N \sim 4.1$ K [1]. Recently, the existence of a unique phase stabilized only at a finite temperature and a finite magnetic field, the so-called "A-phase", has been revealed in this material, and its similarity with the magnetic skyrmion phase of MnSi has attracted much attention [2,3]. Furthermore, recent neutron scattering experiments have revealed that a helical magnetic ground state at zero magnetic field, and in A-phase, a six-fold diffraction pattern in a plane perpendicular to the applied magnetic field, which is characteristic of the skyrmion lattice, was observed [4]. However, the detailed magnetic structure and the temperature and magnetic field dependence of the order parameter were not clarified.

We have studied the behavior of the magnetic order parameter of EuPtSi by resonance X-ray scattering (RXS). RXS is a technique that enables element-selective observation of ordered states of a magnetic element by using resonant X-rays at the absorption edge of atoms. The experiments were performed at the beamline BL22XU of SPring-8 and the beamline BL-3A of Photon Factory using a single crystalline sample prepared by the Bridgman method. As a result of the RXS measurements, a clear resonance signal was observed near the L_2 absorption edge of Eu, and the superlattice magnetic reflections were successfully observed in both the helical phase and the A-phase. From the detailed temperature and magnetic field dependence of these reflections and the presence or absence of higher-order satellite reflections (Fig. 1), it was found that the helical phase is in a single-Q multi-domain state and the A-phase is in a triple-Q state. Since the typical skyrmion lattice is described by the triple-Q order, these results, together with the single-Q helical ground state, suggest the formation of a skyrmion lattice state in EuPtSi.

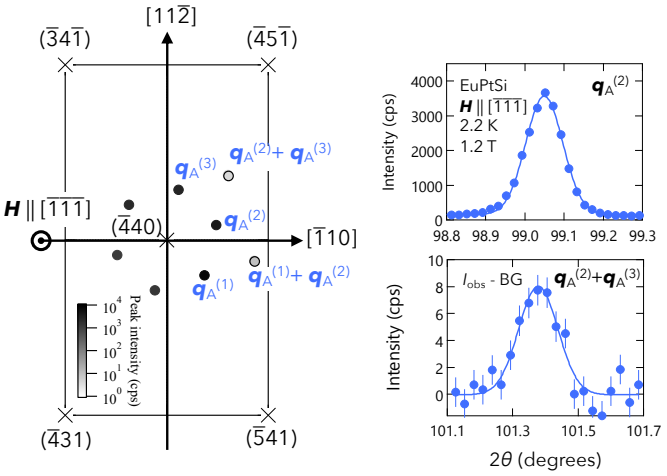

Fig. 1. Diffraction pattern around (-4, 4, 0) observed in the A-phase of EuPtSi (left) and peak profiles of the RXS signals of the first satellite magnetic reflection and the second satellite magnetic reflection (right).

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Probing the A phase of EuPtSi by neutron scattering

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Magnetic skyrmions are particle-like, topologically non-trivial spin textures that have attracted interest because of their fundamental aspects as well as applications due to their potential as a low-power-consumption device. The magnetic skyrmion lattice can be represented by a triple-$\boldsymbol{q}$ magnetic order in which three chiral propagation vectors make an angle of 120˚ with each other in the plane normal to the applied field $\boldsymbol{H}$. This realization was first identified in a chiral magnet MnSi by the observation of the six-fold magnetic pattern in the small angle neutron scattering[1]. As the number of skyrmion compounds increases, variety properties of skyrmions have been unveiled. Unlike this diversity, magnetic ions have been mostly limited to 3*d* transition metal elements.

Recently, high-quality single crystals of the 4*f* electron compound EuPtSi, which belongs to the same space group as MnSi, have been grown. An application of magnetic fields in EuPtSi induces a transition from an antiferromagnetic ground state to a field-induced ordered phase. This induced phase, called *A* phase, has unique characteristics; the *A* phase exists only in the finite temperature-field range[2], where an additional Hall resistivity was observed. This feature, common to the skyrmion phase of MnSi, implies the existence of a magnetic skyrmion in EuPtSi. In order to get microscopic insights into the magnetic ordering phenomena in EuPtSi, neutron diffraction and small-angle neutron scattering experiments on single crystals were carried out[3].

Our experiments revealed that the ground state is a single-chiral helical structure described by the ordering vector $\boldsymbol{q}_1$ = (0.2, 0.3, 0) and its cyclic permutations. When magnetic fields were applied along [1,1,1] to enter into the *A* phase, magnetic peaks are moved into the plane orthogonal to the applied fields, and form 6-fold patterns with keeping periodic length of $\boldsymbol{q}_1$. The observed peaks can be described by the unique ordering vector $\boldsymbol{q}_A$ = (0.09, 0.20, -0.28) and its cyclic permutations with retention of the order of the signs. This result indicates a triple-$\boldsymbol{q}$ magnetic structure in the plane normal to the applied field, that evidences a formation of skyrmion lattice in the *A* phase.

In contrast to similarities in the phase diagram and transport, the *A* phase in EuPtSi has distinct differences from MnSi. One major difference is shorter periodicity by a factor of 1/10 in EuPtSi than MnSi. This originates in a relatively strong coupling to the lattice in EuPtSi, which may give rise to an anisotropic magnetic phase diagram for the *A* phase. Indeed, we confirmed that the triple-$\boldsymbol{q}$ structure is vanished by rotating an applied field direction. The present discovery in the 4*f* electron magnet will open new possibilities to deepen our understanding on skyrimion physics.

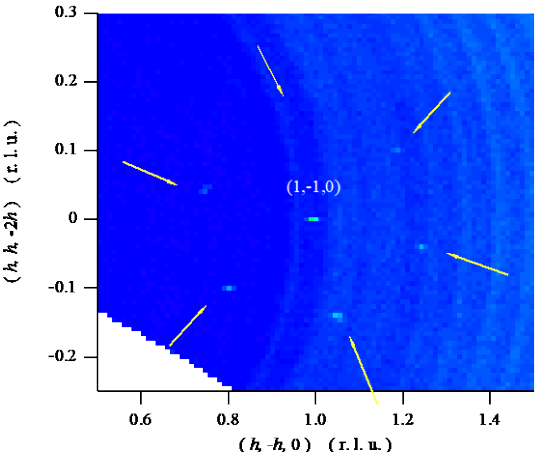

Fig. 1 Neutron scattering intensity map in the A phase of EuPtSi. Hexagonal magnetic patterns were observed around (1, -1, 0).

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First order transition and magnetic fluctuation of EuPtSi observed by ^{151}Eu Mössbauer spectroscopy

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A new type of topological magnetic texture so called the magnetic skyrmion was discovered in the A-phase under the magnetic fields just below the Néel temperature T_N of the B20-type helimagnets such as MnSi and FeGe having the trillium lattice. The trillium lattice is also realized in EuPtSi intermetallic compounds of LaIrSi type structure. Recently, the first-order like antiferromagnetic ordering of EuPtSi at T_N=4.0 K, which is small in magnitude compared with ordering temperatures of 10–100 K in the usual divalent Eu compounds, reflecting the frustration of spins in the chiral structure. Very recently, the magnetic structure of EuPtSi has been investigated by neutron diffraction and resonant x-ray scattering measurements.

We report the results of ^{151}Eu Mössbauer spectroscopy on EuPtSi with a chiral structure belonging to the $P2_13$ (#198) space group at zero magnetic fields.[1] The paramagnetic single absorption forms a magnetic splitting profile directly below T_N=4.0 K and the spectrum at 3.9 K consists of the sum of the paramagnetic single absorption and the magnetic splitting absorption, which indicates a first-order transition at T_N=4.0 K. The temperature dependence of the effective hyperfine fields H_{hf} at the Eu nucleus follows a power law of $H_{hf} = H_0(1-T/T_N)^{\beta}$ with H_0 = 25.6±0.4 T and β = 0.16±0.01 indicates a full moment of Eu^{2+} ions. The relative integral intensity I of the ^{151}Eu Mössbauer spectroscopy for EuPtSi is enhanced using a Debye calculation below $T_0 \simeq 15$ K, which indicates the development of magnetic correlations. The line widthΓ of the ^{151}Eu Mössbauer spectra increases below T_{FD} ($\simeq$8 K) close to the Weiss temperature θ_p and shows a sharp peak at T_N. The magnetic behavior of EuPtSi can be divided into three magnetic regions: the paramagnetic (PM) phase, the fluctuation-disordered (FD) phase and the antiferromagnetic phase with fluctuation disordering (AF with FD). The magnetic fluctuation is realized around the first-order transition between the paramagnetic and helimagnetic states in EuPtSi.

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NMR and μSR studies of EuPtSi

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EuPtSi crystallizes in a non-centrosymmetric chiral structure (the cubic LaIrSi-type) belonging to the same space group $P2_13$, as MnSi and other B20 materials[1,2]. Magnetic frustrations originating from the geometry of the trillium lattice of Eu ions and/or from competing ferromagnetic and antifferromagnetic interactions have been suggested to play an important role in stabilizing the skyrmion lattices in EuPtSi. In zero magnetic field, EuPtSi exhibits a helimagnetic phase transition at T_N = 4 K[3]. The transition is the first-order, and further, has been suggested to involve strong magnetic fluctuations as a precursor to the transition in some temperature range above T_N from the result of the specific heat, the magnetic susceptibility and Mössbauer[2, 4-6]. The magnetization also indicates the existence of a triciritcal point under magnetic fields, suggesting the suppression of magnetic fluctuations by the magnetic fields[5]. From the data, the possibility of a fluctuation-induced first-order transition has been discussed for EuPtSi, while nature of magnetic fluctuations responsible for that had not been well identified.

To investigate to the fluctuation-induced first-order transition, we carried out ^{29}SiNMR and μSR measurement of EuPtSi. The single crystal sample for NMR measurements was prepared with the ^{29}Si isotope in order to enhance the ^{29}Si NMR signal intensity, since the natural abundance of the NMR-active nucleus ^{29}Si is only 4.7%. μSR measurements under zero magnetic field (ZF) were performed at D1 port of J-PARC and General Purpose Surface-Muon Instrument (GPS) of Paul Scherrer Institut.

Both the NMR and μSR measurements have detected the development of magnetic fluctuations of Eu spins in the paramagnetic state far above T_N. However, these fluctuations are suppressed rapidly under magnetic fields. In zero-field, diverging critical fluctuations have been found to emerge in a relatively wide temperature region above T_N as shown in Fig.1. The results support the scenario of the fluctuation-induced first-ordered transition, and further, suggest the existence of large magnetic frustrations in the spin system.

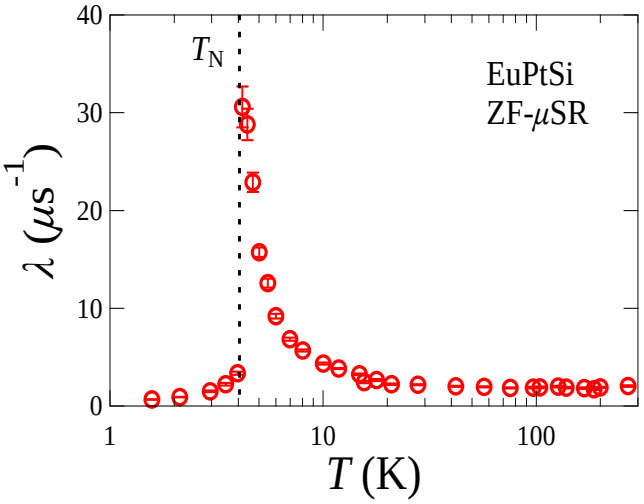

Fig. 1 Temperature dependence of the relaxation ratio λ in zero-magnetic field.

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Magnetochiral Dichroism in Antiferromagnets

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Magnetochiral dichroism (MChD) is a type of nonreciprocal directional dichroism in which optical absorption changes by the reversal of propagation direction of light. In this presentation, we report MChD in two antiferromagnets with cation ordered corundum related structure.

The first one is $MnTiO_3$, which belongs to ilmenite structure with space group $R\bar{3}$. Below Neel temperature T_N = 65 K, Mn^{2+} spin moments antiferromagnetically align along the *c*-axis [1]. Since Mn^{2+} form buckled honeycomb structure in *ab* plane, antifferomagnetic order of Mn^{2+} moments cause violation of space inversion and time reversal symmetry, then the magnetoelectric (ME) effect appears [2]. The magnetic point group of the antiferromagnetic state is $\bar{3}'$, this magnetic point group have antisymmetric components in the off-diagonal terms of ME tensor, these terms related to the toroidal moment. Therefore, we measure MChD in antiferromagnetic $MnTiO_3$, which have toroidal moment in antiferromagnetic phase. As a result, MChD in $MnTiO_3$ was successfully observed in the antiferromagnetic phase without an external magnetic field [3]. The MChD of this material is caused by the toroidal moment of the Mn^{2+} site [4], which depends on the direction of the antiferromagnetic vector of Mn^{2+}.

The second one is Cr-doped Ni_2InSbO_6, which belongs to chiral and polar space group *R*3 [5]. Ni_2InSbO_6 has an antiferromagnetic helical magnetic order below T_N [6]. Substitution of magnetic Cr^{3+} for the non-magnetic In^{3+} of Ni_2InSbO_6 results in a ferrimagnetic helical magnetic structure in which the net magnetic moments do not cancel [7]. Reflecting this ferrimagnetic component and the chiral crystal structure, we found that the MChD in the Cr-doped Ni_2InSbO_6 [8].

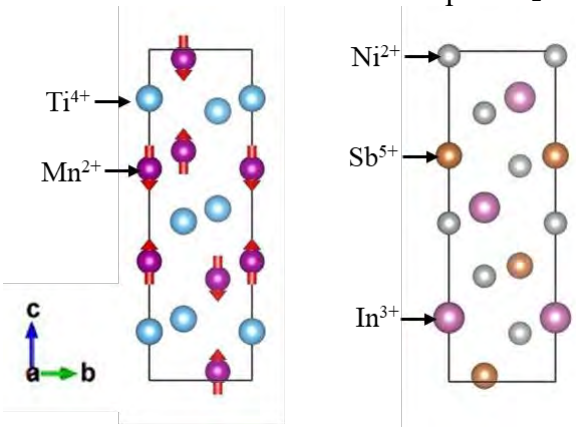

Figure. Cation ordering corundum related structure of (left) $MnTiO_3$ and (right) Ni_2InSbO_6.

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Pressure-induced magnetic phases in $YbNi_3Ga_9$ with a chiral crystal structure: AC-calorimetric and Hall resistivity measurements up to 12 GPa

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Yb-based compounds $YbNi_3X_9$ (X = Al, Ga), crystallizing in the trigonal $ErNi_3Al_9$-type structure with a space group $R32$, are expected to exhibit chiral magnetic structures as well as quantum critical phenomena [1, 2]. $YbNi_3Al_9$ undergoes an antiferromagnetic (AFM) transition at T_M = 3.4 K [3]. By substituting Cu for Ni by 6%, a chiral soliton lattice (CSL) is realized under magnetic fields B applied parallel to the a-axis [1, 4, 5]. While $YbNi_3Ga_9$ is in an intermediate-valence state at ambient pressure, a magnetic order is induced by applying the pressure P above P_c=9 GPa [6]. At 8.6 GPa ≅ P_c, the specific heat C divided by temperature T, C/T, exhibits $-\ln T$ dependence for $2 < T < 9$ K [7]. At $T < 1.5$K, C/T is saturated to a constant value 1 J/K^{2}mol as shown in Fig. 1. For $P \geq 11$ GPa, T_M and B_c increase up to 5 K and 1.3 T, respectively [7], whose values are comparable to those for $Yb(Ni_{0.94}Cu_{0.06})_3Al_9$ with CSL [4]. Furthermore, a field-induced ordered phase (phase II) appears only for $B \perp c$ as shown in Fig. 2 [7].

In order to investigate the feasibility of CSL in the pressure-induced magnetic phases in $YbNi_3Ga_9$, we have measured the magnetoresistance $R(B)$ and the Hall resistivity $\rho_H(B)$ for $B \perp c$ under P up to 12.6 GPa. The slope of $\rho_H(B)$ changes between the phase I and phase II as shown in Fig. 3. This behavior is similar to that observed in $Yb(Ni_{0.94}Cu_{0.06})_3Al_9$ at the phase boundary of B = 1.0 T. Furthermore, the field dependence of $R(B)$ having a maximum near the boundary between phase II and paramagnetic (PM) phase is similar to that for $Yb(Ni_{0.94}Cu_{0.06})_3Al_9$. These close resemblances between the two systems suggest the formation of CSL in the phase I and/or phase II of $YbNi_3Ga_9$.

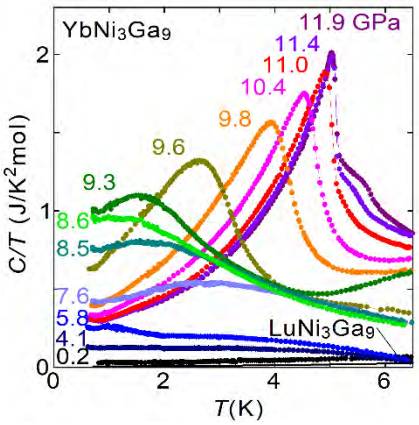

Fig. 1. Temperature dependence of the specific heat of $YbNi_3Ga_9$ under various pressures [7].

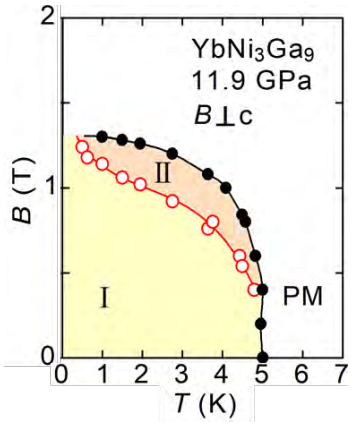

Fig. 2. B-T phase diagram of $YbNi_3Ga_9$ for $B \perp c$ at 11.9 GPa determined by specific-heat measurements [7].

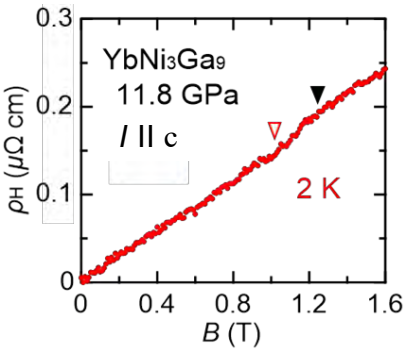

Fig. 3. Hall resistivity $\rho_H(B)$ of $YbNi_3Ga_9$ for $B \perp c$ at 11.8 GPa. The red open arrow and black closed arrow show the fields at the boundaries between phases I and II and phase II and PM phase, respectively.

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Observation of current-induced magnetization corresponding to magnetic point group

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The magnetoelectric (ME) effect is characterized by the appearance of a magnetization under the application of an electrical field or by the appearance of an electrical polarization under the application of a magnetic field. In ME effect, the field and the polarization are connected by a second rank tensor [1]. At present, ME effect has been studied mainly in insulators [2, 3]. On the other hand, current-induced magnetization –magnetocurrent effect– has been reported for metallic UNi_4B and attracts much attention recently [4, 5].

In this study, we focused on the rare-earth intermetallic compound of $ErNi_3Ga_9$. This compound crystallizes in a chiral crystal structure with the space group of *R*32. Below T_N=6.4K, the *Ising*-like antiferromagnetic structure appears with the magnetic space group of *C*2 [6]. The tensor forms of the linear ME effect are $\alpha = \begin{pmatrix} a_{11} & 0 & 0 \\ 0 & a_{22} = a_{11} & 0 \\ 0 & 0 & a_{33} \end{pmatrix}$ and $\begin{pmatrix} a_{11} & 0 & 0 \\ 0 & a_{22} & a_{23} \\ 0 & a_{32} & a_{33} \end{pmatrix}$ for the point group of 32 and 2, respectively [1]. We can expect the appearance of a magnetization $\boldsymbol{M}$ by the application of an electrical current $\boldsymbol{J}$ with $\boldsymbol{M}=\alpha\boldsymbol{J}$ in $ErNi_3Ga_9$.

To clarify electrical current effect on the magnetization in $ErNi_3Ga_9$, we measured the temperature dependence of magnetization while applying the DC electrical current to the crystallographic axis. The magnetization was measured using a commercial SQUID magnetometer (MPMS, Quantum Design, Inc.) in the temperature range of 2-15 K with a magnetic field of up to 100 Oe.

As a result, we observed the rise in magnetization by the electrical current corresponding to the diagonal term of a_{11} and a_{33} below T_N. This means that a_{11} and a_{33} increase with the magnetic ordering. For off-diagonal term of a_{12}, the current-induced magnetization was not detected in the experimental uncertainty. This is consistent with a_{12}=0. We have also tried to detect the off-diagonal response for the term of a_{23}, but not observed the electrical current effect until now.

The detail of current-induced magnetization in $ErNi_3Ga_9$ corresponding to the tensor form will be presented. I will also talk about the achievements and issues of chiral magnetism research in rare-earth intermetallic compound.

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Cross-Correlated Phenomena Driven by Augmented Multipoles

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We have developed the theoretical framework of the augmented multipoles in order to describe an arbitrary electronic degrees of freedom in crystal, and classified a variety of cross-correlated phenomena in terms of the augmented multipoles on the basis of crystallographic point-group symmeties [1,2]. In particular, we have derived the quantum-mechanical expressions of the magnetic and electric toroidal multipoles, which allow us to discuss microscopically the corresponding quantities with relevant wave function of electrons [3].

In this framework, we propose that the electric toroidal quadrupole ordering of the complicated bond modulation (Fig. 1) occurs in the spin-orbit coupled metal $Cd_2Re_2O_7$ [4], which has been elucidated by several experiments at moment. Other application is a generation scheme for magnetic structure basis (Fig. 2) classified according to the symmetry-adopted multipoles [5]. The procedure generates the appropriate basis set to search for stable magnetic structure in combination with first-principle density functional theory calculation. We also show that a class of organic antiferromagnets serve as a spin current generator by a thermal gradient or an electric field, even with vanishing spin-orbit coupling [6]. The mechanism relies on the occurence of the spin splitting in band structure under a collinear antiferromagnetic ordering, whose microscopic conditions are examined by means of augmented multipoles from general point of view [7]. We are further developing a theory on the spin splitting without relying on the atomic spin-orbit coupling including a non-collinear antiferromagnetic ordering [8].

A series of works has been done in collaboration with many co-workers, especially Yuki Yanagi, Satoru Hayami, Megumi Yatsushiro, Yukitoshi Motome, Makoto Naka, Michito Suzuki, Hitoshi Seo, Ryotaro Arita, Takuya Nomoto.

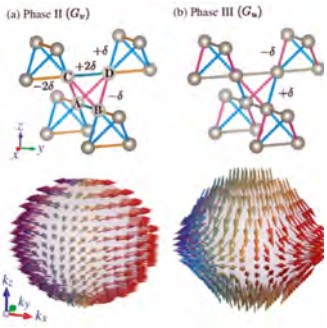

Fig. 1. Order parameter candicates, ETQ, in $Cd_2Re_2O_7$.

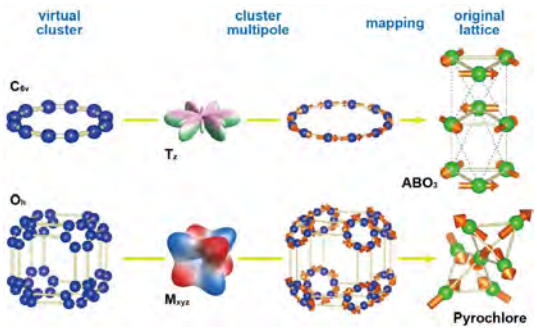

Fig. 2. Generation scheme for magnetic structure basis by symmetry-adopted multipoles.

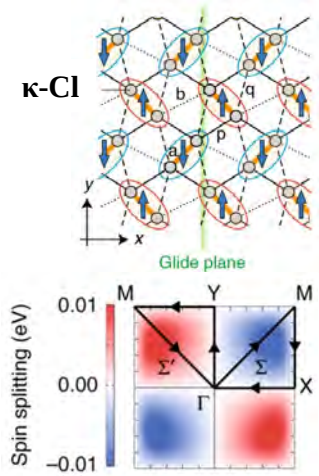

Fig. 3. Spin splitting in κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$.

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Large room-temperature responses induced by cluster magnetic octupoles in the non-collinear antiferromagnet Mn_3Sn

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In the last few years, there has been a surge of interest in antiferromagnetic (AF) materials due to their favorable properties for device applications including a vanishingly small stray field, faster spin dynamics, and more abundance in nature compared to their ferromagnetic counterparts [1]. In fact, motivated by these intriguing properties, several breakthroughs have been made: for example, an anisotropic magnetoresistance (an even-function response under time-reversal (TR)) has been found to be useful for detecting collinear AF ordering [2]. Another breakthrough is the discovery of an odd-function response under TR in the non-collinear antiferromagnetic metal Mn_3Sn such as an anomalous Hall effect (AHE) [3], anomalous Nernst effect (ANE) [4], magneto-optical Kerr effect (MOKE) [5], and novel type of (magnetic) spin Hall effect [6] at zero magnetic field. Moreover, both theoretical and experimental studies have revealed that Mn_3Sn is the first version of a TR symmetry breaking Weyl metal (Weyl magnet) possessing a large and controllable Berry curvature (fictitious field) in momentum space because of its unique magnetic and electronic structures, which contribute to the sizable TR-odd responses such as AHE and ANE [7].

In this talk, we will propose the Weyl antiferromagnet Mn_3Sn as a promising material for the AF spintronics. This is because Mn_3Sn exhibits the large TR-odd responses at room temperature and zero magnetic field [3], and these spontaneous responses can be controlled by the AF spin state with the ferroic ordering of cluster magnetic octupoles [8], which has been recently observed by the MOKE microscopy [5]. We will also show that Mn_3Sn thin films even on the amolphous exhibit the large TR-odd responses at room temperature as well as the bulk Mn_3Sn [9]. These findings provide an important step for the further development of spintronics devices using AF materials.

This work is based on the collaboration with D. B. Gopman, Y. P. Kabanov, R. D. Shull (NIST), O. M. J. van 't Erve (U.S. NRL), L. Wu (UPenn), D. Rees, S. Patankar, J. Orenstein (UCB), T. Koretsune, M.-T. Suzuki (Tohoku Univ.), R. Arita (RIKEN), Y. Li, C. L. Chien (JHU), and Y. Otani (ISSP, Univ. of Tokyo).

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Theoretical Study of anomalous Hall effect and order parameter in antiferromagnets

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Antiferromagnets exhibit manifold physical properties leading to a fascinating play ground in condensed matter physics. In particular, discoveries of a large anomalous Hall (AH) effect [1], anomalous Nernst effect [2], and magneto-optical Kerr effect [3] in noncollinear antiferromagnetic ordered states of Mn_3Sn and Mn_3Ge attracted great attention in recent years. We have extensively investigated the interplay between the macroscopic phenomena and magnetic structures with first-principles calculations combined with the analysis of magnetic symmetry and order parameter of the antiferromagnetic structure. We introduced cluster multipoles as order parameters to characterize the antiferromagnetic order as a natural extension of the magnetic dipole moment of ferromagnetic order. It has been shown that the large AH effects in Mn_3Sn and Mn_3Ge are induced with the symmetry breaking characterized by a cluster magnetic octupole moment that belongs to the same irreducible representation (IR) as that for the magnetic dipole moment, leading to large imbalanced +/− contribution of the Berry curvature to the AH conductivity in ***k***-space [4]. We further proposed a scheme to generate the symmetry adapted complete orthonormal basis set of multipole magnetic structures for arbitrary crystal structure [5] (Fig. 1). These outcomes facilitate the understanding of order parameters in antiferromagnets and provides a missing link between the order parameters and macroscopic phenomena in antiferromagnets.

The generation scheme of symmetry adapted multipole magnetic structures can be applied to the systematic research of magnetic structures such as stable magnetic structure, macroscopic phenomena, and electronic structures by combining it with first-principles calculations. We introduce in the presentation our systematic research to identify topological features which enhance the AH conductivity in the non-collinear antiferromagnetic metallic states of antiperovskite manganese nitrides, Mn_3AN, making use of the multipole expansion for magnetic structures with the first-principles calculations [6]. We show that small Berry curvatures widely spread throughout the Fermi surface in the Brillouin zone, dominantly contribute after the ***k***-space integration to the AH conductivity in Mn_3AN.

These works were supported by JSPS KAKENHI Grant Numbers JP15H05883 (J-Physics), JP15K17713, JP18H04230, and JST PRESTO.

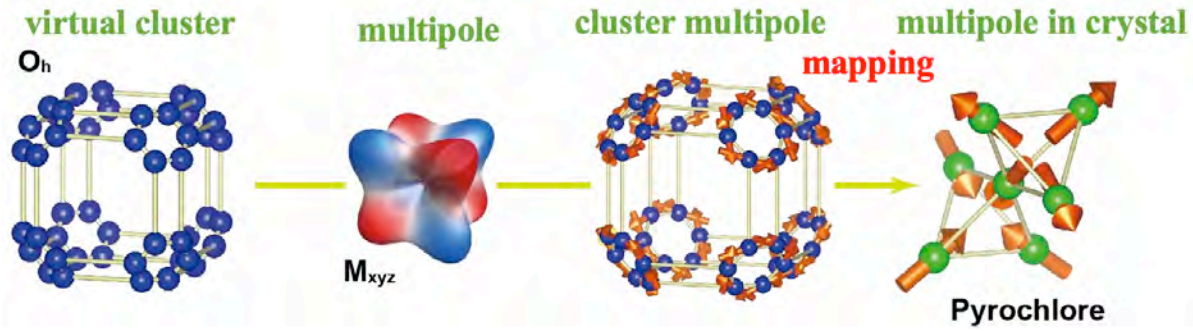

Fig. 1. Procedure to generate multipole magnetic structure in crystal [5].

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Augmented magnetic octupole of Mn_3Sn investigated by x-ray magnetic circular dichroism

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Kagome antiferromagnet Mn_3Sn has attracted great interests, since it exhibits the large anomalous Hall effect [1] and the large anomalous Nernst effect [2] despite the extremely weak ferromagnetic moment (~ $2m\mu_B$ par Mn atom). Then an augmented magnetic octupole formed on the Mn cluster is noted as the origin of these large responses [3]. Moreover, the large magneto-optical Kerr effect (MOKE) originated from the augmented magnetic octupole has been reported [4], although the MOKE was considered to be able to probe the ferromagnetic moment but not the moment in antiferromagnetic state.

In this study, we have investigated the augmented magnetic octupole in Mn_3Sn by utilizing x-ray magnetic circular dichroism (XMCD), although the XMCD technique was considered to detect a net magnetization, like the MOKE. Then we succeeded in measuring a peculiar XMCD spectrum, which cannot be simply explained by the weak ferromagnetic moment in Mn_3Sn. By the theoretical consideration [5], it was elucidated that the XMCD signal is originated from the magnetic dipole operator, the so-called *Tz* term. Moreover, there is a strong link between the augmented magnetic octupole and the *Tz* term. Finally, it suggests that the XMCD can become an effective probe to observe multipole moments in antiferromagnets.

This research was performed in collaboration with M. Kimata, N. Sasabe, Y. Yamasaki, C. Tabata, Y. Yokoyama, Y. Kotani, M. Ikhlas, T. Tomita, H. Nojiri, K. Amemiya, T. Nakamura, and S. Nakatsuji.

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Charge melting induced by magnetic field in BaMn2Pn2 (Pn = As, Sb and Bi)

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A numerous exotic materials intriguingly adopt the tetragonal $ThCr_2Si_2$ (122) crystallo-graphic structure, and more than 400 compounds have been known to be classified into this structure. An extended family related to $BaFe_2As_2$, where high temperature superconductivity can be found by chemically doping or under high pressure, is also categorized into the same family. An intriguing subfamily of $BaMn_2Pn_2$ (Pn: P, As, Sb, and Bi) compounds can be differentiated from the other $BaTM_2Pn_2$'s for their remarkable ground states with special symmetry as well as intriguing transport properties. Antiferromagnetic $BaMn_2Pn_2$ adopts tetragonal (122) layered structure with a G-type antiferromagnetic ordering (AFM) of a Mn^{2+} sublattice. The magnetic point group of this structure is 4'/m'mm', which is odd under both time reversal (TR) and space inversion (SI) symmetry, but the combination symmetry of PT (SIxTR) is preserved. By taking into account the *p-d* hybridization between Mn's and Pn's orbitals, these materials have been predicted to host magnetic multipolar orders that promise interesting physical properties [2]. On the other hand, the half-filled electronic configuration of Mn^{2+} and the *p-d* hybridization also suggest a strongly correlated unconventional ground state [1]. We have recently reported our observations on a new kind of large negative magnetoresistance (LMR) that is exhibited by all itinerant antiferromagnetic BMPn's [3]. Under zero magnetic field (**H**), itinerant antiferromagnetic BMPn's are electrically small band gap semiconductors. The LMR emerged under a large in-plane magnetic field ($\mathbf{H}_{ab}$) reduces the resistivity by more than -98 % and leads to a recovery of a metallic state with the charge carrier melting. Interestingly, this LMR is strongest when **H** is perpendicular to the direction of the magnetic sublattice so that the Mn^{2+} moments are most effectively tilted. We suggest that the LMR can be considered as a consequence of the breaking of the PT symmetry, which is protected by the crystallographic and magnetic structure. However, the mechanism and the physical implications of the LMR still remain unclear. Towards the better understanding of the LMR in itinerant antiferromagnetic BMPn's, we have been carrying out detailed electrical transport and magnetic experiments under high **H** in BMPn's. Progresses will be presented.

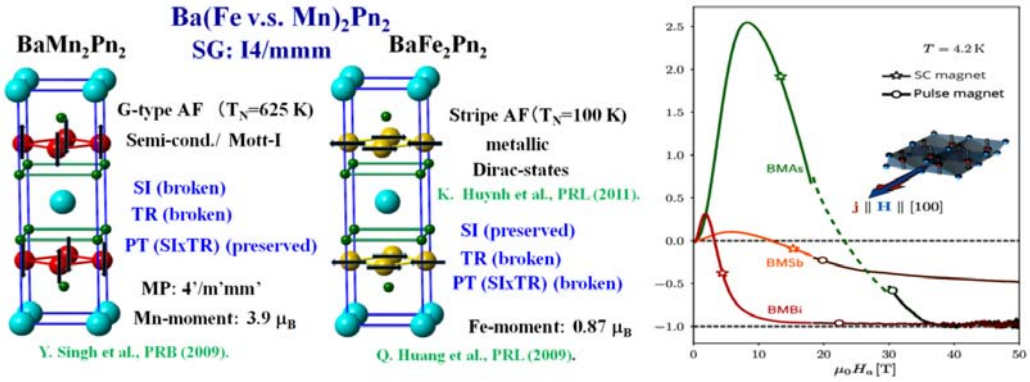

Fig. Structure of $BaMn_2Pn_2$ and observed large magneto resistance (LMR)

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Observation of a magnetopiezoelectric Effect in the antiferromagnetic Metal $EuMnBi_2$

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Magnetopiezoelectric effect, which refers to a linear strain response to electric currents and its inverse response in low-symmetric magnetic metals, is a generalization of magnetoelectric effects in insulators to metals. In metallic materials with high conduction-electron densities, static (dc) piezoelectric responses are not allowed, even if the metals have a symmetry group low enough to support a static polarization. This is because the static surface charge density is screened out by bulk conduction electrons. However, it was recently proposed [1] that dynamic distortion can arise in response to electric currents without screening effects in antiferromagnetic metals that simultaneously break time-reversal and spatial-inversion symmetries.

Here, we have experimentally studied the magnetopiezoelectric effect [1] in the antiferromagnetic metal $EuMnBi_2$ [2,3] (T_N = 315 K). Using laser Doppler vibrometry at low temperatures, we found that dynamic displacements emerge along an inplane direction upon application of ac electric currents in the [001] direction in $EuMnBi_2$ at low temperatures [2,3]. The displacement signals showing up in response to the electric current increase in proportion to the applied electric currents. We confirmed that such displacements are not observed along the [001] direction of $EuMnBi_2$ or $EuZnBi_2$ with nonmagnetic Zn ions, consistent with the symmetry requirement of the magnetopiezoelectric effect [1]. As temperature increases from the lowest temperature, the displacement signals decrease monotonically, showing that magnetopiezoelectric signals are larger for higher conductivity states.

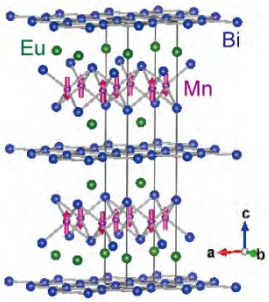

Figure: Crystal structure of $EuMnBi_2$ together with magnetic structure of Mn moments. The antiferromagnetic order of Mn moments (T_N = 315 K) breaks both space inversion and time reversal symmetries, allowing the magnetopiezoelectric effect.

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Multipole degrees of freedom and off-diagonal responses

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In condensed matter physics, multipole moments are often used to describe anisotropic electric charge and electric current distributions. In particular, in *f* electron systems, higher-rank multipoles have been introduced to describe anisotropic electronic order parameters, such as an electric quadrupole and a magnetic octupole [1-3]. Recently, such a concept of multipole has been recognized as important objects to represent various degrees of freedom in electrons, such as charge, spin, and orbital, from the microscopic point of view by including electric toroidal and magnetic toroidal multipoles in addition to conventional electric and magnetic multipoles [4]. The systematic descriptions by using four types of multipoles enable us not only to identify complex exotic order parameters, such as the electric toroidal quadrupoles [5-7] and magnetic toroidal dipoles [8, 9], but also to predict intriguing cross-correlated phenomena including the magneto-electric and magneto-elastic effects [10, 11].

In the present study, we present a general microscopic formalism of multipoles and its extension to cluster, hybrid, and bond multipoles. By showing microscopic multipole expressions in real and momentum space and discussing a classification of multipoles under crystallographic point groups, we demonstrate when and how the four multipole degrees of freedom can become active in physical space. Furthermore, we show how the multipole degrees of freedom affect the electronic band structure and off-diagonal linear responses. For example, the magneto-electric (-current) tensor is characterized by the magnetic monopole, magnetic toroidal dipole, and magnetic quadrupole (electric toroidal monopole, electric dipole, and electric toroidal quadrupole), which becomes finite in the absence (presence) of the time-reversal symmetry. We also discuss the multipole degrees of freedom is deeply related to the emergent spin splitting in the band structure. We show that cluster multipole in a unit of antiferromagnet gives rise to momentum-dependent spin splitting even without the atomic spin-orbit coupling.

This work has been collaborated with Hiroaki Kusunose (Meiji University), Yuki Yanagi (Tohoku University), and Megumi Yatsushiro (Hokkaido University) and was supported by JSPS KAKENHI Grants No. JP18H04296 (J-Physics)..

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Current-induced magnetization in the antiferromagnetic ordered state of Ce_3TiBi_5

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We found a new antiferromagnetic (AFM) heavy fermion compound Ce_3TiBi_5 with AFM ordering temperature of T_N = 5.0 K. Ce_3TiBi_5 has a hexagonal structure ($P6_3/mcm$). Ce ions form a zig-zag chain structure parallel to the *c*-axis. An inversion center exists at the midpoint of the nearest Ce-Ce bond but not at the Ce site. We focus on the lack of the local inversion symmetry at the Ce site. A theoretical study has predicted that magnetization can be induced by an electric current in the AFM ordering on the zig-zag chain. Therefore, Ce_3TiBi_5 is an attractive candidate for studying the ME effect in the sense of the augmented multipole.

In this study, we investigate the magnetoelectric (ME) effect of the AFM compound Ce_3TiBi_5. We carried out magnetization measurements on Ce_3TiBi_5 under an applied constant electric current and a static magnetic field around the AFM ordering temperature. We have additionally tried to find new compounds with the same crystal structure of Ce_3TiBi_5. Moreover, we have tried to develop a new method to measure the current-induced magnetization under low temperature and high pressure.

We successfully observed of the current-induced magnetization below T_N. The magnitude of the current-induced magnetization has a linear electric current dependence and exhibits no magnetic field dependence. This behavior is consistent with the theoretical prediction of the ME effect in the ferrotoroidal ordered state. Some new compounds of RE_3TiBi_5 are found and identified their crystal structure to be isostructural with Ce_3TiBi_5, where RE = La, Ce, Pr, Nd, Sm, and Gd. However, magnetoelectric effect of these compounds without Ce_3TiBi_5 were not observed yet.

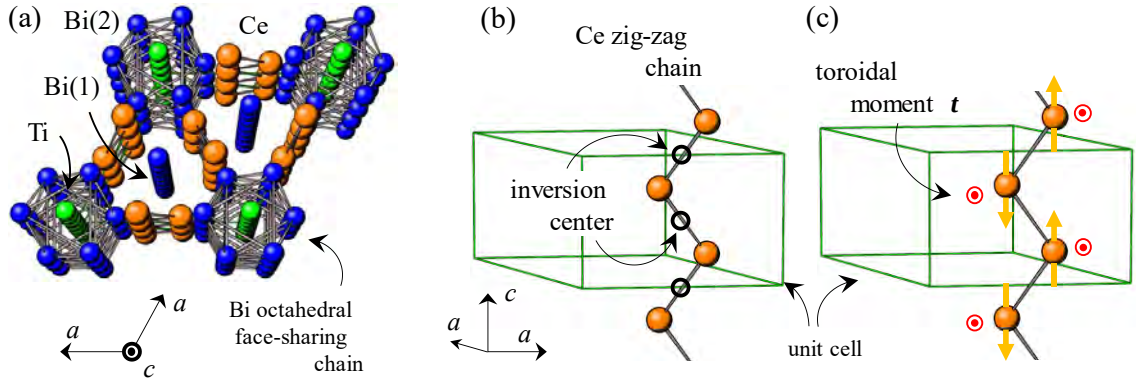

Fig. 1. (a) Top view of the crystal structure of Ce_3TiBi_5 (space group $P6_3/mcm$). (b) Schematic side view of the crystal structure, where only Ce ions on a front side are drawn for clarity. Green line denotes a unit cell. Black circles indicate the position of the inversion center. (c) Magnetic structure on the Ce zig-zag chain, where the unit cell does not change even at the ordered state. Orange arrows on the Ce ions indicate ordered magnetic moments below T_N, which is expected from the anisotropy of $\chi(T)$. The red mark denotes a toroidal moment on a Ce ion.

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Transport properties in $Cd_2Re_2O_7$

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$Cd_2Re_2O_7$ shows two structural transitions at T_{s1} = 200 K and T_{s2} = 120 K. These transitions are considered as the band Jahn-Teller effect due to the Fermi surface instability in the semi-metallic band structure. [1] $Cd_2Re_2O_7$ transforms to the superconducting state at T_c = 1.0 K. The *s*-wave superconductivity at ambient pressure is experimentally established in spite of the lack of the inversion symmetry below T_{s1}. [2] Application of pressure suppresses the T_{s1} transition at P_c = 4.2 GPa. [3] While the T_c increases twofold with increasing pressure, the upper critical field B_{c2} also increases and reaches 7.8 T near P_c, which is 27 times larger than that at ambient pressure. Recently, Fu and his coworkers theoretically suggested the parity-fluctuation scenario where the fluctuations of the parity-breaking order generate an attractive pairing interaction in an odd-parity pairing channel. [4] However, the electronic band structure in the ground states is not clarified experimentally even in the ambient pressure due to the presence of the multi domain.

In this study, we carried out the Hall effect measurement under high pressure up to 4.7 GPa in order to investigate the electronic states in the pressure-induced phases and search the fluctuation effect of the parity-breaking order. At ambient pressure, the Hall coefficient R_H shows the positive value in phase I and the negative value in phase II, and finally larger negative value in phase III, which agrees with the previous result [1] except the low temperature part. (Fig. 1) In the pressure-induced phases IV, VII and VIII, R_H shows larger negative values, which is similar to the pressure dependence of the effective mass in the previous report [3]. (Fig. 2) This behavior seems to be related to the hole-band splitting around the K-point, predicted by the band calculation. [1] The hole-band splitting causes the heavier effective mass of holes due to the phonon correction of the band dispersion near the Fermi level. Therefore, the smaller mobility of holes compared to the electrons leads to the negative R_H. The enhanced phonon contribution in the pressure-induced phases may be the fluctuation effect of the parity-breaking order.

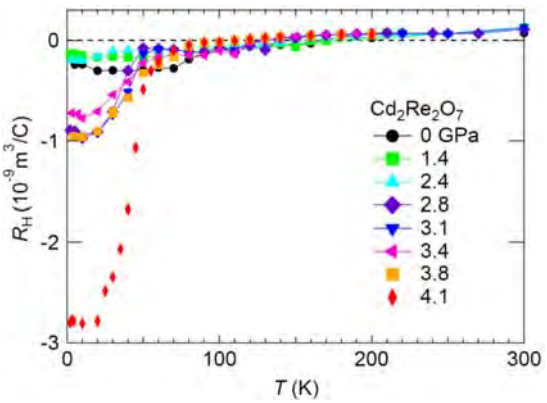

Fig. 1. Temperature dependence of the Hall coefficient in $Cd_2Re_2O_7$ under high pressure.

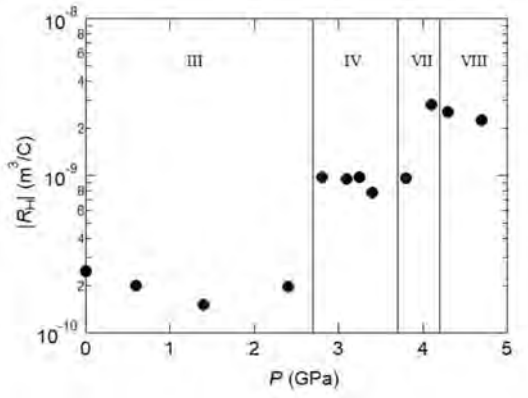

Fig. 2. Pressure dependence of the Hall coefficient at 10 K in $Cd_2Re_2O_7$.

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Electric toroidal quadrupoles in spin-orbit coupled metal $Cd_2Re_2O_7$

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Spontaneous inversion symmetry breaking in electron systems with strong spin-orbit coupling has attracted growing interest [1]. In these systems, many intriguing physical phenomena, such as the spin-momentum locking, magnetoelectric effect, and nonreciprocal response, emerge due to the antisymmetric spin-orbit coupling activated by the electronic orderings [2,3]. Pyrochlore oxide $Cd_2Re_2O_7$ is an archetypal spin-orbit coupled metal exhibiting spontaneous inversion symmetry breaking [4,5]. Upon changing the temperature and pressure, this compound shows rich electronic phases including the several parity-broken states [6]. In addition, among many pyrochlores, it is the only superconductor thus far [4]. The superconducting state also shows unconventional behavior under pressure, presumably due to the spontaneous parity breaking [6,7].

At ambient pressure, $Cd_2Re_2O_7$ shows two successive structural transitions at T_{s1}~200K and T_{s2}~120K. For $T>T_{s1}$ (phase I), the space group is Fd-3m symmetry while for $T<T_{s1}$, that is lowered to the tetragonal one lacking the inversion symmetry [5]. Since the observed lattice deformation at the transition is very small, the phase transition is expected to be electronic origin. However, the space group symmetry for $T<T_{s1}$ is still controversial. According to the X-ray diffraction experiments [5], the space group is I-4m2 for $T_{s2}<T<T_{s1}$ (phase II) and $I4_122$ for $T<T_{s2}$ (phase III). On the other hand, the nonlinear optical measurements imply that the (magnetic) space group is I-4 (I-4m'2' or I-4'm'2) in the time-reversal-preserving (-breaking) scenario [8,9].

In this study, we theoretically investigate the potential order parameters for the phases II and III in $Cd_2Re_2O_7$ [10]. We propose that the symmetry-breaking states in the phases II and III are characterized by electric toroidal quadrupoles with E_u symmetry. By performing a microscopic analysis based on a tight-binding model, we show that the electric toroidal quadrupoles are activated by spontaneous modulation of the hopping amplitudes and/or the spin-current ordering (see Fig. 1). We also discuss a way to detect the electric toroidal quadrupoles by investigating the characteristic physical properties in the ordered states, such as the Edelstein effect and nonreciprocal transport.

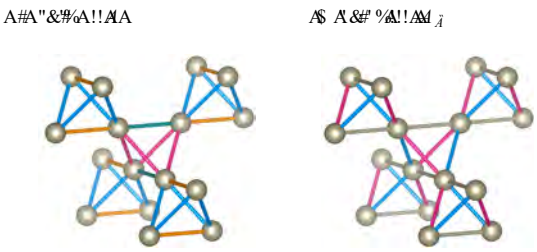

Fig. 1. Schematic pictures of the bond modulations by the electric toroidal quadrupoles (a) for phase II and (b) for phase III.

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Material development of layered Bi-based superconductors

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Since the discovery of BiS_2-based superconductors with a typical crystal structure displayed in Fig. 1a, we have studied physical and chemical characteristics of BiS_2-based superconductors [1]. Recent theoretical and experimental investigations have suggested unconventional pairing mechanisms [2]. Furthermore, possible nematic superconductivity has been observed in $LaO_{0.5}F_{0.5}BiSSe$ [3]. Based on those notable characteristics and the similarities of layered structure and physical properties to cuprates and Fe-based superconductors, further understanding of the superconductivity of layered Bi-based compounds has been desired.

Recently, we have developed four-layer-type Bi-based superconductors whose crystal structure is displayed in Fig. 1c. Bulk superconductivity was induced by multi-site substitution in $La_2O_2Bi_3(Ag,Sn)(S,Se)_6$ and $(La,Eu)_2O_2Bi_3(Ag,Sn)S_6$. The chemical pressure effects are useful for the emergence of bulk superconductivity, which is a trend like the typical BiS_2-based (two-layer-type) systems [4-6]. In addition, one-layer-type superconductor (Fig. 1a) was also synthesized by Ruan et al. [7]. In the presentation, we will talk about physical and chemical properties of those layered Bi-based superconductors and discuss about the similarities and differences among them.

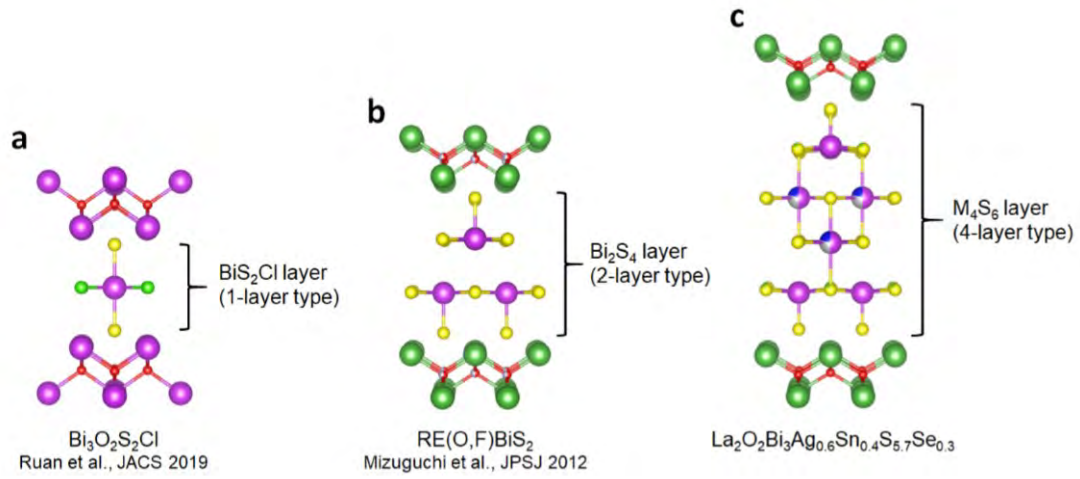

Fig. 1. Schematic images of crystal structure of (a) one-layer-type, (b) two-layer-type, and (c) four-layer-type Bi-based layered superconductors.

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Materials Development of p-/f-electron Systems

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The electronic properties of the rare earth elements are typically dominated by the +3 oxidation state. Well-established exceptions include Ce, Sm, Eu, Tm, and Yb compounds, in which the +4 or +2 oxidation states can be also stabilized. Of particular interest in such circumstances are selected rare earth systems (Kondo insulators, heavy fermions) for which the 4f levels lie close to the Fermi level, E_F and may exhibit mixed valence phenomena. Unambiguous signatures of electronically-driven valence changes with changes in external stimuli (temperature, pressure) are found for instance in the variation of the elastic, electronic, and magnetic properties of such mixed valence solids. Here, we focus on emergent electronic phenomena in a family of hybrid f-/p-electron molecular materials. In particular, we have synthesized and researched new families of molecular-based strongly correlated f-electron fullerides in which the presence of the electronically-active C_{60} anions is combined with mixed configuration rare earth ions potentially leading to properties intrinsically unattainable in other currently available systems. Strong correlations dominate the electronic properties of both the rare-earth cation and the C_{60} anion sublattices. To-date our work has unambiguously led to the authentication of the occurrence of valence transitions with novel characteristics arising from the simultaneous presence of the electronically-active C_{60} sublattice.

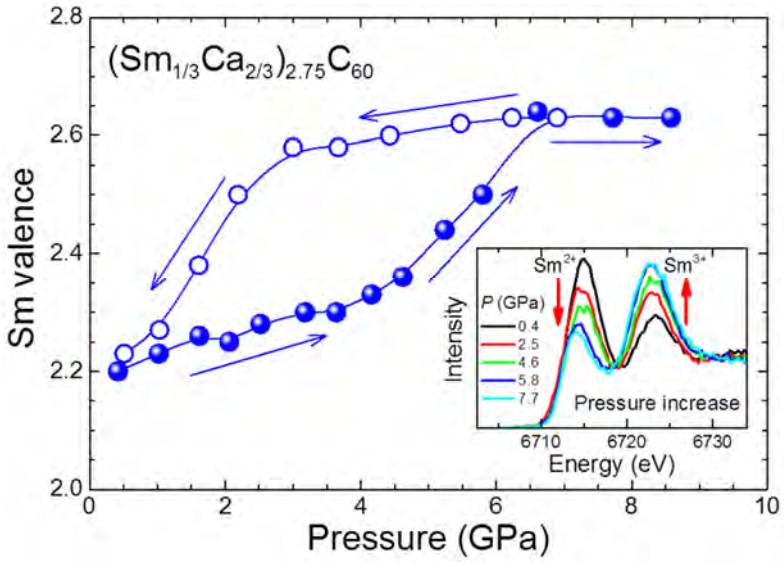

Fig. 1. Evolution of Sm valence in $(Sm_{⅓}Ca_{⅔})_{2.75}C_{60}$ with changes in applied pressure. Inset: Partial fluorescence yield-X-ray absorption (PFY-XAS) spectra upon both pressurization.

Novel Phenomena in Geometrically Frustrated Iridates

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Studies on the physical properties of Ir oxides for the last ten years have revealed the importance of spin-orbit interaction (SOI). The purpose of our project to reveal novel phenomena in geometrically frustrated iridates of $Ln_2Ir_2O_7$ and $Ca_5Ir_3O_{12}$. We will report our recent results.

(I) Pyrochlore iridates $Ln_2Ir_2O_7$ (Fd-3m, No. 227) for Ln=Nd - Ho indicate metal-insulator transition (MIT) [1]. The MIT involves AFM state with Ir moments with "all-in/all-out" configuration which is considered to be octupole ordering [2]. We have investigated the effect of chemical carrier doping for pyrochlore iridate $Y_2Ir_2O_7$ (T_{MI} = 165 K) in order to reveal the theoretically predicted QCP [3,4]. The resistivity of hole doped $Y_2Ir_2O_7$ shows upward convex T dependence around disappearance of AFM; the number of powers of T dependence in the resistivity is 0.69 for Cu3% and Ca25% below 40 K down to 2.2 K. This upward convex T dependence in the resistivity is observed only in hole doped $Y_2Ir_2O_7$ as far as we have examined.

(II) $Ca_5Ir_3O_{12}$ with hexagonal structure with noncentrosymmetric space group of P-62m (No. 189) shows semiconducting behavior [5]. In the crystal structure, one-dimensional chains of the edge-sharing IrO_6 form triangular lattices in the c-plane. $Ca_5Ir_3O_{12}$ has a mixed valance state of Ir^{4+} and Ir^{5+} and the averaged valence of Ir ions is +4.67. $Ca_5Ir_3O_{12}$ indicates a second order phase transition at 105 K and AFM ordering below T_N =7.8 K [5]. However, below 105 K, there is no evidence of structural transition in XRD and ND experiments [5]; this phase transition at 105 K is "*hidden order*". Furthermore, $Ca_5Ir_3O_{12}$ shows nonlinear electrical conductivity along the c-axis in the non-ordered state [6]. We have investigated the nonlinear conductivity and the origin of phase transition at 105 K. First, we performed harmonic voltage response experiments by using AC current. Consequently, odd harmonics were observed below 200 K which is a proof of nonlinear conductivity [7]. Next, we obtained the strong evidence that the phase transition at 105 K is a structural phase transition by Raman scattering [8]. All observed peaks are assign by using the energy of phonon mode at Γ point calculated by GGA with SOI (SO-GGA). New peaks appear below 105 K; theses modes are related to vibration by oxygen [8]. Next, we obtained the phonon dispersion by using inelastic X-ray scattering. Then, we found that the phonon dispersion calculated by SO-GGA is in very good agreement with experimental result [9]. Furthermore, the low-energy phonon properties such as the Debye temperature, specific heat, and sound velocity is also good agreement with the calculation by SO-GGA.

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Regular-Triangle Trimer Formation in the Metal Insulator Transition of β-Pyrochlore Oxide CsW_2O_6

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Metal-insulator transitions in *d* electron systems with pyrochlore structure have been a longstanding issue in the field of condensed matter physics since the discovery of Verwey transition in magnetite. They are caused by various mechanisms such as a dimer formation in $CuIr_2S_4$ and $LiRh_2O_4$ and a ferroic order of extended magnetic octapoles in $Cd_2Os_2O_7$ and $Nd_2Ir_2O_7$. Here we report that the β-pyrochlore oxide CsW_2O_6, where $W^{5.5+}$ ions having $5d^{0.5}$ electron configuration form a pyrochlore structure, exhibits a unique electronic phase transition different from other pyrochlore systems. CsW_2O_6 was first synthesized by Cava *et al*. in 1993 [1] and then found to show a metal-insulator transition accompanied by a decrease of magnetic susceptibility [2]. However, formation mechanism of the transition is still not fully understood, because all the previous studies were employed using polycrystalline samples or thin films [3].

We succeeded in preparing single crystals of CsW_2O_6 by a vapor-phase growth method. As shown in Fig. 1(a), electrical resistivity of CsW_2O_6 single crystals strongly increases at T_{MI} = 215 K with decreasing temperature. Magnetic susceptibility also decreases at this transition, consistent with the previous results obtained by using polycrystalline samples [2]. Single crystal X-ray diffraction experiments indicated that a structural change from *Fd*–3*m* to $P2_13$ space groups preserving the cubic symmetry occurs at the 215 K transition [Fig. 1(b)]. Below 215 K, charge order with non-integer valance and ordering of 5*d* orbitals confine all 5*d* electrons to W_3 trimers with an equilateral triangular shape. Electronic instability appeared in the band structure of *Fd*–3*m* phase is suggested to play an important role for the formation of a W_3 trimer.

The work has been mainly done in collaboration with K. Niki, R. Mitoka, Y. Yokoyama, K. Takenaka, H. Amano, N. Katayama, H. Sawa, Y. Nakamura, H. Kishida (Nagoya University), H. Harima (Kobe University), T. Hasegawa, N. Ogita (Hiroshima University), Y. Tanaka, M. Takigawa (University of Tokyo), K. Takehana, Y. Imanaka (NIMS).

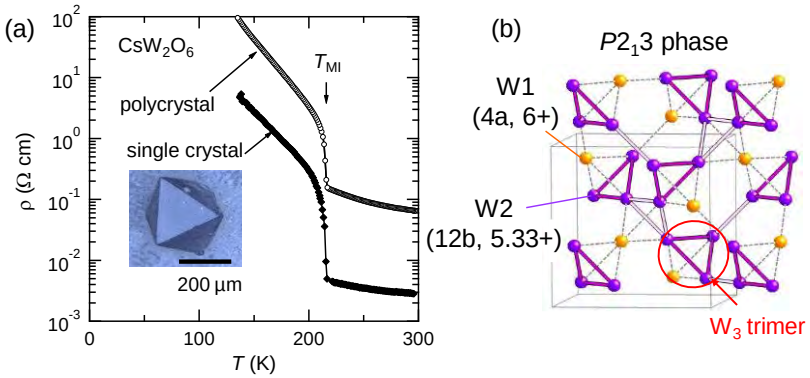

Figure 1: (a) Temperature dependence of electrical resistivity of single crystals and polycrystalline samples of CsW_2O_6. (b) W sublattice in the $P2_13$ phase.

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Control of physical properties in Ni and Pd chalcogenides

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The transition-metal chalcogenides MX_2 (M=transition metal, X=S, Se, Te) have several crystal structures such as 2-dimensional CdI_2-type, pseudo-2-dimensional $PdSe_2$-type and 3-dimensional pyrite-type structures. In this work, we have studied MX_2 (M=Ni and Pd) and related systems to explore the new physical properties. These systems show various physical properties coupled with the crystal structures.

First targets of our study are $PdSe_2$ and doped compounds. $PdSe_2$ has pseudo 2-dimensional $PdSe_2$-type structure, and undergoes the structural phase transition from $PdSe_2$-type to 3-dimensional pyrite structure by applying pressure. The previous study on $PdSe_2$ using pressure has indicated that this material shows the superconductivity with T_c~13 K in the high-pressure pyrite phase [1]. In our work, we have tried to control this structural phase transition under the ambient pressure by Ni- and Rh-doping to $PdSe_2$. $NiSe_2$ and $RhSe_2$ are famous pyrite-type materials, and the solid solution systems of $Pd_{1-x}Ni_xSe_2$ and $Pd_{1-x}Rh_xSe_2$ with pyrite structure can be synthesized by the high pressure method [2,3]. We have succeeded in the synthesis of polycrystalline samples of pyrite-type $Pd_{1-x}Ni_xSe_2$ and $Pd_{1-x}Rh_xSe_2$ (x>0.3) under high pressure (~5 GPa). These Ni- and Rh-doped compounds with x=0.3~1.0 show the metallic behaviour indicating the semiconductor-metal transition concomitantly with structural phase transition from $PdSe_2$ to pyrite around x~0.3. Only in the Rh-doped system, we have observed the superconducting transition in the narrow region of doping level (x~0.8). This superconductivity may be related with that of $PdSe_2$ under hydrostatic pressure.

Second target of our work is $NiTe_2$. The previous experimental studies on the related systems $PdTe_2$ and $PtTe_2$ with CdI_2–type layered structure have indicated that these compounds have type II Dirac fermion state [4,5]. The results of angle resolved photoemission spectroscopy (ARPES) have revealed that the Dirac point exists at $(0,0,k_z)$ and the Dirac cone is strongly tilted along Γ-A direction (k_z-direction) in $PdTe_2$ and $PtTe_2$. Our ARPES result for $NiTe_2$ indicates that this compound also has type II Dirac cone. The Dirac point exists very near Fermi level (E_F) in $NiTe_2$, while they in Pd and Pt system are located around -0.5 ~ -0.8 eV below E_F. We also measured ARPES for Pd-doped $NiTe_2$ system $Ni_{1-x}Pd_xTe_2$. The present result of ARPES has indicated that the energy level of Dirac point gradually decreases with Pd doping level.

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Search for π-d system in organic molecular conductors

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We have been searching for spin-charge complex electron systems in organic molecular conductors to explore new spin-charge coupled phenomena. In organic molecular conductors, (BEDT-TTF)$_2X$ (X: monovalent anion) salts are good candidates, which show a wide variety of electronic phases including antiferromagnetic and weak ferromagnetic states. However, most of (BEDT-TTF)$_2X$ salts do not show distinct spin-charge couplings, even in the multiferroic κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl with weak ferromagnetic and ferroelectric states [1]. Although this material does not show any anomaly in the dielectric constant under magnetic field [1], it was found that the charge and spin degrees of freedoms are well coupled to the lattice sector [2]. Thus, π spin-charge couplings and/or another spin system is one of the most important issue of organic materials.

Our target is α''-(BEDT-TTF)$_2$Rb$_{1.2}$Co(SCN)$_4$ which shows an anomalous increase in Hall coefficient in contrast to the similar charge-ordered θ-(BEDT-TTF)$_2$(Rb, Cs)(Co, Zn)(SCN)$_4$ salts. In addition, the magnetocapacitance effect shows cusp like anomaly (Fig. 1(c)), indicating d spin and π spin-charge coupling. To demonstrate the π-d spin-charge couplings in this material, we performed infrared optical spectroscopy measurements under magnetic field to demonstrate the correspondence with the magnetocapacitance anomaly. Organic materials, in general, are so small that far-infrared spectra cannot be obtained without using synchrotron radiation. Magneto-optical station at BL43-IR in SPring-8 is suitable facility for our purpose, where we have improved the light intensity and usable wavelength for several years [3].

Figure 1(a) and 1(b) shows the infrared optical conductivity and reflectivity spectra of α''-(BEDT-TTF)$_2$Rb$_{1.2}$Co(SCN)$_4$ at 10 K under magnetic field ($H \parallel a^*$), respectively. The main spectral shape does not change by the increase in magnetic field, but small deviations can be discerned, where the reflectivity increases at lower wavenumbers in contrast to the higher ones. This tendency can be clearly represented by the center of wavenumber for optical conductivity from 500 to 1250 cm^{-1}, $\omega^* = \int \omega \cdot \sigma(\omega)\, d\omega / \int \sigma(\omega)\, d\omega$ shown in Fig. 1(d). The change in ω^* corresponds well with the anomalies in the magnetocapacitance effect probably due to spin flops (Fig. 1(c)). This result indicates the strong charge-spin coupling of this system.

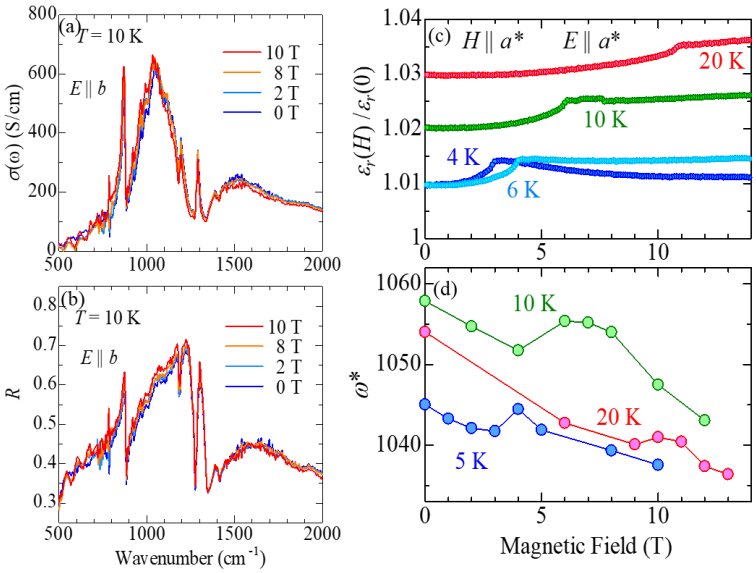

Fig. 1 Infrared optical (a) conductivity and (b) reflectivity at 10 K. Magnetic field-dependence of (c) dielectric constant and (d) the center of wavenumber for optical conductivity.

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Development of novel non-Kramers doublet compounds and its multipole properties

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Cubic Pr-based compounds PrT_2X_{20} (T: Transition metal, X = Zn, Al) have been intensively studied because they exhibit a variety of fascinating phenomena such as a pressure induced superconductivity, and non-Fermi liquid behavior [1-6]. It has been suggested that these phenomena arise from the multipolar degrees of freedom of the non-Kramers doublet crystalline electric field (CEF) ground state.

Recently, we have succeeded in synthesis of new cubic Pr-based compounds $PrRu_2X_2Zn_{18}$ (X = Sn and In), which are ordered derivatives of PrT_2Zn_{20} [7, 8]. The magnetization and specific heat measurements revealed that the CEF ground state of $PrRu_2X_2Zn_{18}$ (X = Sn and In) is a non-Kramers doublet [7, 8]. For $PrRu_2Sn_2Zn_{18}$, a phase transition was observed at T_o = 1.9 K by electrical resistivity and specific heat measurements. The magnetic entropy at T_o is close to Rln2, indicating that the phase transition is attributed to the multipolar degrees of freedom of the non-Kramers doublet. In addition, we found that the electrical resistivity of $PrRu_2Sn_2Zn_{18}$ shows a shoulder structure at around 20 K, which is possibly attributed to the interaction between 4*f* and conduction electrons. In the presentation, we will also discuss the low temperature properties of $PrRu_2In_2Zn_{18}$.

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Itinerant multipolar order in spin-orbit-coupled metals

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Novel quantum phases emerging in the presence of strong spin-orbit interactions (SOIs) are attracting great attention in condensed matter physics. An itinerant electron system with a large SOIs possesses a unique Fermi liquid instability, which drives a phase transition to a variety of odd-parity multipolar ordered phases accompanied by a spontaneous space inversion symmetry breaking [1]. The 5*d* pyrochlore oxide $Cd_2Re_2O_7$ (CRO) is considered as a promising candidate hosting this novel phase transition. Space inversion symmetry in the room-temperature structure of $Cd_2Re_2O_7$ is lost across the phase transition at $T_{s1} \sim 200$ K [2]. A very recent theoretical work pointed out that electric toroidal quadrupole (ETQ) orders are realized in this compound below T_{s1} [3]. In this ETQ ordered phase, antisymmetric SOI lifts the spin degeneracy of Fermi surface at the high-temperature phase. The resultant spin-split Fermi surfaces are predicted to induce specific electromagnetic responses depending on the created ETQ order pattern.

In order to observe the ETQ order and detect the expected electromagnetic responses, we grew high-quality and reasonably-large crystals of $Cd_2Re_2O_7$ and performed various physical properties measurements. Our continuous effort to improve sample quality has resulted in high-quality single crystals of sub-millimeter size with the residual resistivity ratio higher than 400, which is one order of magnitude higher than those of previously prepared crystals. We also established the way to align the tetragonal twin domains formed below T_{s1} by using a piezo stack, as shown in the Fig. (a). This technique enables us to reveal the intrinsic properties of $Cd_2Re_2O_7$, such as anisotropy of resistivity, for the first time [Fig. (b)]. Attempt to observe the electromagnetic responses is ongoing.

In addition to the detailed study on $Cd_2Re_2O_7$, we investigated multipolar orders of spin-orbit-entangled 5*d* electrons. The results on the Mott insulating double perovskite Ba_2MgReO_6, in which quadrupolar order is observed [4], and the metallic 5*d* oxide $PbRe_2O_6$, where successive phase transitions accompanied by a large reduction in the density of states [5], will be presented.

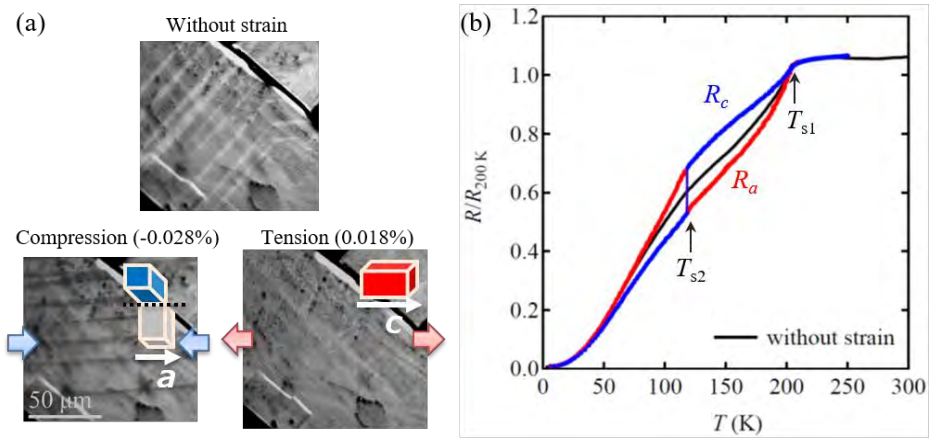

Figure (a) Polarizing microscope images of a pristine (1 0 0) surface of a $Cd_2Re_2O_7$ crystal. Three kinds of twin domains observed without strain is merged into single domain under tensile strain. (b) The temperature dependence of the anisotropy of resistivity for $Cd_2Re_2O_7$ measured under compressive and tensile strains. R_a and R_c represent the resistivity along the *a* and *c* axis, respectively.

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Observations of Fermi surfaces of Conductive Multipole Materials

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The rare earth intermetallic compounds RT_2Al_{20} (R = rare-earth, T = Ti, V, Cr) with cubic $CeCr_2Al_{20}$-type structure (space group $Fd\bar{3}m$, No. 227) have attracted much attention because of their fascinating unusual properties, such as the heavy-fermion superconductivity in the proximity of multipole (or quadrupole) order in PrT_2Al_{20} [1-4], Nd-based heavy-fermion behavior in ferromagnetic NdV_2Al_{20} [5], and the field-insensitive heavy-fermion behavior in $SmTi_2Al_{20}$ [6, 7]. Also, Laves-phase compound $CeIr_2$ with cubic $MgCu_2$-type structure (space group $Fd\bar{3}m$, No. 227) has attracted much attention because of the interesting physical properties, such as the mixed valent and the heavy-fermion superconductivity [8, 9]. Recently, this compound also attracted much attention for photocathode material because of the high quantum efficiency [10]. To understand both the physical properties and the origin of good performance for the application, it is essential to study these electronic states.

During this project, we have investigated the Fermi surface properties in RTi_2Al_{20} (R = La, Pr, Nd, and Sm) and RIr_2 (R = La and Ce) by means of the de Haas-van Alphen (dHvA) effect and the band structure calculation. In the former compounds for R = Pr, Nd, and Sm, the features of the angular dependences of the dHvA frequencies are close to that of the reference compound $LaTi_2Al_{20}$, indicating the localized nature of 4*f* electrons in these compounds, while the magnitudes of mass enhancements are different; for $PrTi_2Al_{20}$ and $SmTi_2Al_{20}$, the cyclotron effective masses m_c^*s are 5 ~ 6 times larger than those of corresponding ones in $LaTi_2Al_{20}$, while for $NdTi_2Al_{20}$, m_c^*s are only about two times larger than those of corresponding ones in $LaTi_2Al_{20}$. These differences should be originated from the differences between these magnetic ground states and/or the strength of *c-f* hybridization. In the latter compounds, we have succeeded in growing good single crystals and observed the dHvA effect for the first time. An almost spherical Fermi surface with the dHvA frequency F = (646 ~ 686) T and the cyclotron effective mass m_c^* = (0.70 ~ 0.82) m_0 (m_0 is the rest mass of an electron) were observed in $CeIr_2$, which is reasonably well explained by the band structure calculation. The feature of Fermi surface in $CeIr_2$ is highly different from that of $LaIr_2$, indicating the 4*f*-electorons in $CeIr_2$ contribute to the conduction bands. Compared to the results of the band structure calculation, relatively heavy mass electrons of 12 m_0 are expected, that contribute to superconductivity below T_c ~ 0.2 K.

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High Magnetic Field Phase of URu_2Si_2-Rh Doping and Temperature Dependent Magnetic Wave Vector

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URu_2Si_2 is one of the most studied U-based strongly correlated electron systems for its rich phase diagram including superconductivity, non-Fermi liquid state, the Hidden Order (HO), high magnetic field-long period antiferromagnetic states and parasitic antiferromagnetic order. A high magnetic field drives the successive phase transition from the HO state to multiple magnetic ordered states named phases II, III and IV. Recently, Knafo *et al*. found that the main wave vector of the field induced ordered phase is incommensurate **k** = (0.6 0 0) through the pulsed high magnetic field neutron diffraction experiment with a reactor neutron source [1]. Because of the absence of the higher ordered satellites, the most plausible order is a spin-density wave (SDW) with sinusoidal modulation. However, the result of the nuclear magnetic resonance suggests deviation from a simple SDW state[2].

On the other hands, in the higher Rh-doped samples of 4 % and 8 %, it was found that the magnetic orders are commensurate **k** = (2/3 0 0). These results show that the so-called phase II are different for non-doped sample and highly doped sample. More recently, the result of Rh 2% sample shows that the phase II is incommensurate and the wave vector is different from the pure sample[3]. By the Rh doping, the system shows the phase transition from incommensurate to commensurate states. It is also notable that suppression of the HO phase at zero field is associated with this transition. Moreover, the disappearance of the (1 0 0) parasitic antiferromagnetic phase within the HO phase has the coincidence with the transition.

We also examined the neutron diffraction measurement under the pulsed high magnetic fields up 37.5 T by time-of-fright spectrometer MERLIN in ISIS spallation neutron source. The combination of the white beam of the spallation neutron and the pulsed field allows us to scan the wide area of the reciprocal space. At 4.2 K, an incommensurate **k** = (0.6 0 0) peak is observed at 37.5 T. It is noteworthy that the linewidth of the (0.6 0 0) peak is only $\Delta h \sim 0.016$, which is about 30 % of the previously reported result [1]. In addition, the small peaks at (0.565 0 0) and (0.625 0 0) are observed. It should be noted that the distribution is along the a*-axis and the temperature is higher than that of [1]. The result shows that the SDW phase at low temperature has multi-q components at higher temperature, though the state is named as a single phase II. These results shows that there are competitions among HO, incommensurate SDW of different wave vectors, commensurate ferrimagnetic like state and the antiferromagnetic state in URu_2Si_2 by temperature variation and by doping. It is instructive that all magnetic modulation is along a*-axis, with which we infer that the anomaly of the Fermi surface along this axis is the key factor for the successive magnetic phase transition.

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Recent Advances in UTe_2

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The discovery of superconductivity in UTe2 attract much attention[1-21]. We present our recent results on the novel spin-triplet superconductor UTe_2, which is at the verge of the ferromagnetic order. The huge upper critical field exceeding the Pauli limit indicates the spin-triplet state. For the field along b-axis, the field reentrant superconductivity is observed up to H_m~35T, where the first order metamagnetic transition occurs. The field reentrant superconductivity in UTe_2 is similar to that observed in ferromagnetic superconductors, namely URhGe and UCoGe. Applying the pressure in UTe_2, the superconducting transition temperature splits and the mulitple superconducting phase is detected in the AC calorimetry measurements. We overview the results on UTe_2 comparing with ferromagnetic superconductors, and show our perspective. This work was done in collaboration with G. Knebel, A. Pourret, J.P. Brison, D. Braithwaite, Q. Niu, M. Valiska, J. Flouquet, A. Nakamura, F. Honda, Y. Homma, D.X,Li, Y. Shimizu, Y. J. Sato, A. Miyake, S. Imajo, S. Kohama, M. Tokunaga, H. Harima, W. Knafo, I. Sheikin, C. Paulsen, Y. Tokunaga, S. Kambe, H. Sakai, S. Fujimori, K. Ishida, G. Nakamine, S. Kitagawa

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Knight-Shift Measurement on U-based compounds and Sr_2RuO_4

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During 5 years of this project, we have been performed NMR measurements on various U-based compounds of conventional *s*-wave superconductor UCo_6, spin-singlet antiferromagnetic (AFM) superconductor UPd_2Al_3, ferromagnetic (FM) superconductor UCoGe, and recently discovered UTe_2[1,2].

In U_6Co, although clear coherence peak just below T_c and an exponential decrease in the superconducting (SC) state were observed in the $1/T_1$ measurement, indicating a conventional full-gap superconductor[3]. The decrease of the Knight shift in the SC state is small and mainly arises from the SC diamagnetic effect. The negligibly small Knight-shift decrease in the SC state is consistent with the absence of the Pauli-limiting behavior in the SC upper critical field H_{c2}. It was found that there are large T independent Van-Vleck-like susceptibility and the spin susceptibility related to superconductivity is small in U_6Co.

In spin-singlet AFM UPd_2Al_3, clear decrease of the Knight shift was observed in the SC state, in good agreement with the previous measurements[4]. The decrease of the Knight shift in the SC state can be well understood with the decrease of the electronic-term in the specific heat measurement below T_c. In addition, it was found that the evaluated values of the Pauli-limiting field H_{Pauli} by using the following relations of $H_{\mathrm{Pauli}} = H_c/\sqrt{4\pi\delta\chi}$ and $\delta\chi = (N_A\mu_B/A_{\mathrm{hf}})\delta K$ are consistent with the experimental values of H_{c2}, showing that H_{c2} are determined by the Pauli-limiting effect. Here, H_c is the critical field of superconductivity, $\delta\chi$ and δK are the decrease of susceptibility and Knight shift in the SC state, and N_A, μ_B and A_{hf} are the Avogadro's number, Bohr magneton and hyperfine coupling constant, respectively. Recently, the anisotropic Knight-shift decrease was found in the SC state of URu_2Si_2[5]. The experimental results are well understood by the above analyses, and indicate that URu_2Si_2 is a spin-singlet superconductor.

In UCoGe, Knight shift was measured in the pressure-induced paramagnetic SC state, where FM order is completely suppressed[6]. We also measured the SC Knight-shift in UTe_2[7]. The results are similar to each other. NMR spectra became broader but hardly shifted across T_c. The Knight-shift change determined from fitting the spectral peak was much smaller than the spin part of the Knight shift estimated from the electronic term in the specific heat measurement. This is sharp contrast with the case of spin-singlet superconductors, but in agreement with the spin-triplet pairing suggested from the large upper critical field. The observed spectrum broadening in the SC state cannot be attributed to the SC diamagnetic effect but would be related to the properties of spin-triplet superconductivity.

We are now investigating the origin of the spectrum broadening in the SC state of UTe_2.

We will also present the recent results of the re-measurements of ^{17}O-NMR Knight-shift Sr_2RuO_4, showing the decrease of the spin susceptibility in the SC state. This measurement was performed, inspired by the recent result by Prof. S. Brown's Group[9].

These measurements were done in the collaboration with K. Deguchi, N. K. Sato, T. Yamamura, D. Aoki, A. Nakamura, F. Honda, Y. Homma, D.X, Li, Y. Shimizu, Y. Tokunaga, S. Kambe, H. Sakai.

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NMR study of URu_2Si_2, URhGe and UTe_2

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We present our recent researches on U-based unconventional superconductors URu_2Si_2, URhGe and UTe_2 by means of NMR. For URu_2Si_2, possible space groups of hidden ordering, and corresponding multipolar order parameters are discussed [1-4]. The Knight shift is consistent with the chiral *d*-wave state [basis function: $k_Z(k_X + ik_Y)$] [5,6]. For URhGe, a strong correlation between the ferromagnetic superconductivity and Ising type magnetic fluctuations [7,8]. In addition, the difference between original and re-entrant superconductivity is addressed. Finally for UTe_2, the peculiar anisotropic and ω-dependent magnetic fluctuation is discussed [9].

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Magnetic fluctuation near the ferromagnetic critical point in UGe_2

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Unconventional superconductivity often appears in the vicinity of magnetic quantum critical point (QCP). However, ferromagnetic (FM) superconductor UGe_2 is an exception.[1] Superconductivity in UGe_2 is enhanced at a 1st-order phase boundary between two kinds of FM states; FM1 and FM2, as shown in Fig. 1. This phase boundary changes to a crossover at high temperatures, that is, the system possesses a critical point (CP) at a finite temperature, which is completely different from the QCP picture. In this circumstance, the superconductivity is realized only in the FM state, and coexists with FM state with the large spontaneous magnetic moment of 0.9 μ_B. Because of these specific features, the SC mechanism in UGe_2 is still under debate. We investigated the microscopic electronic state in the vicinity of the CP using NMR techniques to address the SC mechanism in UGe_2.

In our previous NMR study,[2] we measured nuclear-spin lattice relaxation rate $1/T_1$ under pressure. We successfully observed microscopic coexistence of superconductivity and the FM state, but there is no clear anomaly in the vicinity of the CP, although the critical fluctuation is generally expected there. This is because $1/T_1$ is dominated by the transverse fluctuations. In the FM state, the nuclear quantization axis is coerced to be along the *a* axis, which is the easy axis, because the large internal field is induced. $1/T_1$ corresponds to the magnetic fluctuations perpendicular to the nuclear quantitation axis, that is, the magnetic fluctuations along the *bc* plane. They are the transverse fluctuations. Instead, $1/T_2$ can detect the longitudinal fluctuations, and recently we found the enhancement of $1/T_2$ in the vicinity of the phase boundary, as shown in Fig. 2. This clearly suggests that the longitudinal magnetic fluctuations develop in the region where superconductivity is optimized. This will be a key ingredient to understand the SC mechanism in UGe_2 and the overall picture of FM superconductors.

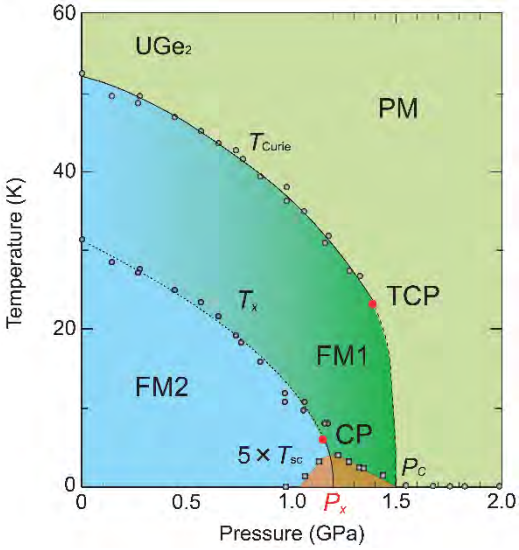

Figure1: *P*-*T* phase diagram of UGe_2

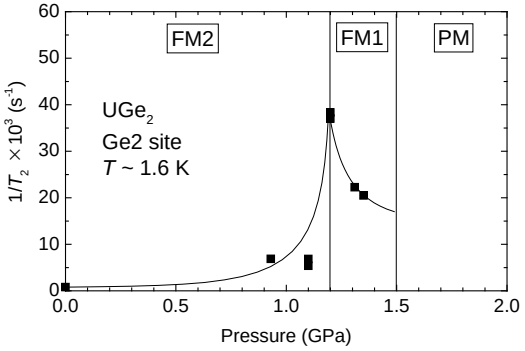

Figure2: *P* dependence of $1/T_2$

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Topology and symmetry of superconductivity in UCoGe and UTe_2

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We theoretically study superconductivity in UTe_2 [1], which is a recently-discovered strong candidate for an odd-parity spin-triplet superconductor [2]. Theoretical studies for this compound faced difficulty because first-principles calculations predict an insulating electronic state, incompatible with superconducting instability. To overcome this problem, we take into account electron correlation effects by a GGA+U method and show the insulator-metal transition by Coulomb interaction [3]. Using the low energy electronic structures obtained as a function of U, we clarify topological properties of possible superconducting states. Fermi surface formulas for the three-dimensional winding number and three two-dimensional Z_2 numbers indicate topological superconductivity at an intermediate U for all the odd-parity pairing symmetry in the I*mmm* space group. Symmetry and topology of superconducting gap node are analyzed and the gap structure of UTe_2 is predicted. Topologically protected low-energy excitations are highlighted, and experiments by bulk and surface probes are proposed to link Fermi surfaces and pairing symmetry. Based on the results, we also discuss multiple superconducting phases under magnetic fields, which were implied by recent experiments.

For UCoGe, we show realization of the *Möbius topological superconductivity* [3,4], which is a topological superconducting phase protected by nonsymmorphic glide symmetry. Interestingly, three of possible four pairing states are characterized by Z_4 topological index. Therefore, UCoGe is a first candidate material for Z_4 topological superconductor which has not been proposed even theoretically.

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Searching for a second superconducting transition in Sr_2RuO_4

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Sr_2RuO_4 has been regarded for many years as a good candidate for a spin-triplet superconductor with the chiral *p*-wave type [1] primarily because NMR Knight shift and spin-polarized neutron scattering experiments have suggested an invariant spin susceptibility across T_c in any field direction [2,3]. However, this conclusion is controversial because an abrupt change in the spin susceptibility for the in-plane H_{c2} has been suggested by low-temperature magnetization measurements [4]. More importantly, in 2019, a substantial reduction in the NMR Knight shift in the superconducting state was detected at low radio frequency pulse powers [5,6]. An analogous *H*–*T* phase diagram accompanied by a first-order phase transition and a Knight-shift reduction has also been established for uniaxially strained samples with $T_c \sim 3.4$ K [5,7]. These results strongly suggest that the first-order phase transition in $H \parallel ab$ is due to the Pauli paramagnetic effect, in contrast to the longstanding chiral *p*-wave scenario.

Thus, an interesting question arises as to whether the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state is realized in Sr_2RuO_4. Indeed, the occurrence of a second transition near H_{c2} for $H \parallel ab$ has been considered for Sr_2RuO_4 in connection with a chiral triplet pairing viewpoint, i.e., splitting of degenerate order parameters in the chiral p_x+ip_y state. This second transition should be carefully examined in high-resolution experiments by using high-quality single crystals showing a first-order phase transition for the in-plane H_{c2}; if a second transition is indeed observed, the origin should be considered in terms of the Pauli-limiting viewpoint.

In our recent report [8], a fourfold oscillation was observed for the low-temperature specific heat under an in-plane rotating magnetic field. In the low-field region, this oscillation can be attributed to quasiparticle excitations around horizontal line nodes with a large in-plane anisotropy in the Fermi velocity. It should be noted that, with increasing *H*, the oscillation was suddenly suppressed at approximately 1.3 T and increased drastically above 1.4 T with an opposite sign. These anomalous behaviors in high fields might be due to the field-angle anisotropy of the FFLO state. In order to search for a possible FFLO phase transition, we have recently developed a ultra-high-resolution dilatometer and performed high-resolution magnetostriction measurements on a high-quality single crystal of Sr_2RuO_4. A first-order phase transition with a field hysteresis was clearly detected below 0.8 K under an in-plane magnetic field. Furthermore, an anomalous hump was found within the hysteresis region below 0.6 K. These findings suggest that Sr_2RuO_4 is a new member of FFLO superconductors.

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Topological superconductors and Weyl superconductors in heavy fermion systems

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There are many candidate materials of topological superconductors and Weyl superconductors in heavy fermion systems. For instance, URu_2Si_2, UCoGe, URhGe, UPt_3 are candidate materials for Weyl superconductors. Also, it is proposed that in Th-doped UBe_{13}, a topological superconducting state, a Dirac superconducting state, and a Weyl superconducting state are realized depending on the Th-concentration and temperature. We, here, report our recent results on unique electromagnetic and thermal transport properties charactering Weyl or topological superconducting states. One of the important features of Weyl superconductors is chiral anomaly of Weyl fermions realized as nodal excitations form point-like gap-nodes. Chiral anomaly results in various interesting transport phenomena such as negative thermal magnetoresistivity [1], chiral torsional magnetic effect [2], and anomalous thermal Hall effect, which is also deeply related to surface chiral Majorana arc states. These chiral anomaly phenomena in Weyl superconductors are induced not only by usual electromagnetic fields and temperature gradient, but also by chiral electromagnetic fields which are generated by lattice strain or topological textures of the superconducting order parameter [3]. The emergence of the chiral electromagnetic responses is also another characteristic feature of Weyl fermions. While the Weyl superconducting state is characterized by bulk transport properties as mentioned above, the experimental verification of the topological superconducting state is more subtle, because most of experimentally observable characteristic phenomena are associated with surface Majorana states rather than bulk properties. However, for some of candidate materials of topological superconductors, the promising pairing states are spin-triplet ones expressed by higher-dimensional representations of point group. For such systems, nematic pairing states which spontaneously break crystal symmetry can be realized. The nematic superconductivity can be experimentally probed by characteristic electromagnetic responses due to collective modes of the nematic superconducting order parameter [4]. We discuss the application of this scenario to Th-doped UBe_{13}.

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Kondo effect and superconductivity in systems with electron and hole Fermi surfaces

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The mechanism of unconventional superconductivities is an important issue in designing a guiding principle to find new superconductors. The heavy electron materials with nearly localized f electrons are typical systems showing unconventional superconductivity, and the identification of the mechanisms has still remained open question since the discovery of $CeCu_2Si_2$ [1] and UBe_{13} [2] in lanthanide- and actinide-based materials. Recently, specific heat measurements in a rotational magnetic field have revealed the full-gap nature of the superconducting states in $CeCu_2Si_2$ [3] and UBe_{13} [4]. This is in contrast with the conventional notion that the strongly correlated electrons favor a spatially non-local and anisotropic pairing. Hence, it is desirable to identify a new mechanism for full-gap superconductivity that is specific to heavy-electron materials.

Motivated by these experiments, we have proposed a mechanism of superconductivity for the Kondo lattice which has compensated-metal conduction bands with electron and hole Fermi surfaces [5]. Here the localized f-electron degrees of freedom are regarded as non-Kramers doublet located at U or Pr atom. This system tends to resolve a frustration associated with the two-channel Kondo effect at low temperatures and results in a quantum mechanically entangled state composed of the Kondo singlet with electron surface and that with hole surface, to break the U(1) gauge symmetry. We clarify the responses to the external electromagnetic fields in Meissner effect [5] and in pair breaking effect by Zeeman splitting [6], which are different from the conventional BCS superconductors.

We also seek for a characteristic feature of non-uniform systems realized in junction systems and quantum vortex states [7]. The length scale of the Cooper pair in our model is very short, which is the order of lattice constant at low energies even when the superconducting gap is small enough. This in strong contrast to the BCS superconductors where the length is simply determined by $\hbar v_F/\Delta$. This is a consequence of the characteristic anomalous self-energy (or pair potential) structure proportional to the inverse of energy. We will discuss this point with the concrete setup.

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Progress on Itinerant multipole and multipole superconductors

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We have studied multipole ordering/fluctuations and unconventional superconductivity emerging in heavy-electron materials, based on the first-principles approach. For examples, rank-5 E^- ordering in URu_2Si_2 [1], d_{x2-y2}-wave pairing state in $CeCoIn_5$ [2], $s_\pm$ -wave pairing state mediated by octupole fluctuations in $CeCu_2Si_2$ [3], and the E_{2u} gap structure in UPt_3 [4]. Then, we have realized that in the multi-orbital superconductors, the superconducting gap functions can be classified like multipole moments. We summarized in tables possible gap functions in typical crystal structures [5]. In addition, we demonstrated that *s*-wave pairing states in multi-orbital superconductors can possess not symmetry-protected but inevitable line nodes or gap minima. As a prototypical example, we studied a gap anisotropy in the BiS_2 layered superconductors in a multi-orbital attractive Hubbard model [6].

Here we will show the obtained phase diagram, and discuss possibility of a non-trivial surface state in d*-wave states. Also, I will introduce the progress on the LDA+DMFT approach.

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Effect of pressure on superconductivity, heavy-fermion and non-Fermi liquid states in Pr1-2-20 compounds

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The cubic Pr-based 1-2-20 compounds with non-Kramers Γ_3 doublet ground state are known as a prototypical system for study the role of multipole degrees of freedom. In particular, PrT_2Zn_{20} (T = Rh, Ir) and PrT_2Al_{20} (T = Ti, V) exhibit a quadrupole order and superconductivity [1-3]. We previously reported that $PrTi_2Al_{20}$ shows the striking enhancement of both the superconducting transition temperature and the upper critical field toward P_c, where the quadrupole order is suppressed by pressure [4]. Further measurements in our new experimental setup for AC-calorimetry using a RuO_2 chip resistors as a heater and thermometer in 10 GPa-class opposed anvil cell, confirmed the pressure evolution of the heavy fermion superconductivity. Above P_c, resistivity shows the non-Fermi liquid (NFL) behavior in wide pressure and magnetic field range, which is in sharp contrast to conventional magnetic quantum critical behavior. It is predicted that strong hybridization between quadrupole moments and conduction electrons leads to the quadrupole Kondo effect, which give rises to the NFL state. Indeed, the temperature dependence of resistivity is well fitted by the quadrupole Kondo lattice model. As a further test for the hybridization effect between the ground doublet and conduction electrons, we have also investigated the effect of pressure on $PrRu_2Sn_2Zn_{18}$, in which the Zn atoms at the 16c site in $PrRu_2Zn_{20}$ are almost preferentially substituted with Sn atoms. $PrRu_2Sn_2Zn_{18}$ does not show the structural transition as observed in $PrRu_2Zn_{20}$ and exhibits a quadrupole order at low temperature [5]. With application of pressure, we found striking enhancement of the overall resistivity, indicating that pressure promotes hybridization analog to the effect of pressure on $PrTi_2Al_{20}$. In this presentation, we will discuss a generic behavior in the Pr-based 1-2-20 compounds under a strong hybridization.

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Single-site non-Fermi liquid behaviors in Pr-diluted 1-2-20 systems with $4f^2$ configuration

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There has been considerable interest in cubic praseodymium-based compounds with non-Kramers doublet ground states of $4f^2$ configuration, because they show a variety of phenomena originating from quadrupolar degrees of freedom. In PrT_2Zn_{20} (T = Rh and Ir) and PrT_2Al_{20} (T = Ti and V), coexistence of quadrupole order and superconducting state was observed, suggesting that the quadrupole fluctuations may play a role in formation of the superconducting pair [1]. Moreover, non-Fermi liquid (NFL) behaviors of the specific heat C and electrical resistivity ρ were observed in PrT_2Zn_{20} (T = Rh and Ir), which indidates a possible formation of the quadrupole Kondo lattice [2-4]. If this is the case, single-site quadrupole Kondo effect could manifest itself when the Pr^{3+} ions are diluted with elements without $4f$ electrons. The single-site quadrupole Kondo effect was first proposed by Cox to interpret the NFL behaviors in UBe_{13} [5]. Theoretical calculations with the two-channel (quadrupole) Kondo model described the magnetic specific heat divided by temperature $C_m/T \propto -\ln T$, quadrupole susceptibility $\chi_Q \propto -\ln T$, and electrical resistivity $\Delta\rho \propto \sqrt{T}$, and presence of the residual entropy of $(1/2)R\ln 2$ at $T = 0$ [6]. Although the quadrupole Kondo effect has been discussed so far in some diluted U and Pr systems, it has not been established yet. In the present work, we have measured C, ρ and χ of diluted Pr^{3+} systems $Y_{1-x}Pr_xT_2Zn_{20}$ (T = Ir, Co) for $x \leq 0.5$ [7,8].

For the Pr diluted $Y_{1-x}Pr_xIr_2Zn_{20}$ with $x = 0.024$ and 0.044, NFL behaviors of $\Delta\rho \propto \sqrt{T}$ and $C_m/T \propto -\ln T$ variations were observed for $0.08 < T < 0.3$ K. The data of the differential electrical resistivity $\Delta\rho$ and C_m/T for $x \leq 0.44$ normalized by the values at the characteristic temperature T_0, which is defined as a temperature that the magnetic entropy reaches $(3/4)R\ln 2$ [6], are well scaled as a function of (T/T_0) for $0.5 \leq T/T_0 \leq 3$. The common characteristics of the NFL behaviors were also observed in the isostructural $Y_{1-x}Pr_xCo_2Zn_{20}$ for $x \leq 0.48$, indicating that the single-site NFL behaviors are independent of not only the Pr concentration x but also the T elements of Co and Ir. In the scaling plots, the C_m/T data of all the samples follow $-\ln(T/T_0)$ for $0.5 \leq T/T_0 \leq 1$. Moreover, those for $x \leq 0.05$ continuously increase down to $T/T_0 = 0.25$ following the $\ln(T/T_0)$ form. On the other hand, the $\Delta\rho$ data for T = Ir of $x > 0.02$ and T = Co of $x > 0.1$, which are divided by the values at T_0, coincide well at $0.25 < T/T_0 \leq 3$. These temperature variations of C_m/T and $\Delta\rho$ are almost the same as those predicted by the single quadrupole Kondo effect with the strong coupling limit [6,9]. Recently, the elastic constant of the $(C_{11}-C_{12})/2$ mode, which corresponds to the Γ_3-symmetry quadrupole susceptibility χ_Q, of $Y_{1-x}Pr_xIr_2Zn_{20}$ for $x = 0.034$ was measured, and the $-\ln T$ variation of $(C_{11}-C_{12})/2$ was observed in the low magnetic fields of $B \leq 2$ T below 0.3 K, confirming the manifestation of the single-site quadrupole Kondo effect [10].

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Elastic Properties of Pr-Diluted Pr1-2-20 systems and $Cd_2Re_2O_7$

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Ultrasonic investigations of the single-site quadrupolar Kondo effect in diluted Pr system $Y_{0.966}Pr_{0.034}Ir_2Zn_{20}$, and the lattice distortion regarding the successive phase transitions in $Cd_2Re_2O_7$ are reported. Since both compounds crystallize in the same space group ($Fd\bar{3}m$, O_7^h, No. 227), it is worthwhile to compare the elastic properties of these compounds by using ultrasound in order to check the possible universality of the Γ_3 (E)-type lattice instabilities and its origin beyond the difference of the electron systems. The elastic constant $(C_{11}-C_{12})/2$ is measured using ultrasonic phase comparative method for the dilute systems $Y_{1-x}Pr_xIr_2Zn_{20}$ ($x = 0$ and 0.034) and $Cd_2Re_2O_7$ single crystals. Brief summaries are as follows :

i) $Y_{1-x}Pr_xIr_2Zn_{20}$

The diluted Pr system of the cubic Pr-based non-Kramers doublet system, $Y_{1-x}Pr_xIr_2Zn_{20}$, has recently been studied systematically. Pr-diluted systems display possible single-site quadrupolar Kondo behavior, which appears as non-Fermi-liquid (NFL) behavior in specific heat and electrical resistivity [1]. We found that the elastic constant $(C_{11}-C_{12})/2$ of the Pr-dilute $x = 0.034$ system exhibits a logarithmic temperature dependence below $T_0 \sim 0.3$ K, where the NFL behavior is observed [2]. This logarithmic temperature variation manifested in the Γ_3-symmetry quadrupolar susceptibility is consistent with the theoretical prediction of the quadrupolar Kondo effect by D. L. Cox [3]. On the other hand, the pure compound YIr_2Zn_{20} without 4*f*-electron contributions shows nearly no change in its elastic constants evidencing negligible phonon contributions for the dilute limit of this system.

ii) $Cd_2Re_2O_7$

The pyrochlore oxide $Cd_2Re_2O_7$ shows two successive structural phase transitions at $T_{s1} \sim 203$ K and $T_{s2} \sim 112$ K [4]. The primary order parameters are theoretically predicted to be a G_u and G_v-type electric toroidal (ET) quadrupoles with Γ_3 (E_u) symmetry for T_{s1} (phase II) and T_{s2} (phase III), respectively [5]. In order to check the even-parity ferro-quadrupolar moments O_u and O_v with Γ_3 (E_g) symmetry, which are possibly induced by these odd-parity ET quadrupoles, we measured the elastic constants $(C_{11}-C_{12})/2$ and C_{44} by using shear ultrasonic modes in wide temperature and magnetic field range. We found that the elastic constant $(C_{11}-C_{12})/2$ shows Curie-type softening from 230 K to T_{s1}, and increasing with cooling down with a strong ultrasonic attenuation in the Phase II ($T < T_{s1}$), while C_{44} mode with Γ_5 (T_{2g}) symmetry shows shoulder like behavior at T_{s1} and keep decreasing toward T_{s2} in phase II. Such clear contrast between these two transverse ultrasonic modes is consistent with the calculated strain susceptibility based on the Landau theory [6]. However, the elastic responses at around T_{S2}, we observed right now, are not consistent with this theory, further investigations, such as ultrasonic measurements under uniaxial pressure to control the possible domains, are now in progress.

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Probing crystalline-electric-field ground-state symmetry by linear dichroism in core-level photoemission

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Ground-state (GS) orbital symmetry determined by the local effective crystalline electric fields (CEF) in strongly correlated electron systems play crucial roles in their functional properties such as superconductivity and multipolar ordering. We have found that the CEF-split GS 4f-orbital symmetry can be probed by linear dichroism (LD) in angle-resolved core d-level hard x-ray photoemission owing to the selection rules in the optical process [1]. We have applied this technique to Ce, Pr, Sm, Dy, Er and Yb-based heavy fermion systems [2–10].

Here we show the linearly polarized angle-resolved Ce $3d_{5/2}$ core-level photoemission spectra and their LDs of tetragonal $CeAgSb_2$ and $CeCu_2Ge_2$ [7,10] in Fig. 1 as the examples of our results. The qualitative difference of LD in the rare-earth $3d_{5/2}$ core-level photoemission spectra in terms of the sign of LD reflects the mutually different CEF-split GS 4f-orbital symmetry, where $CeCu_2Ge_2$ is in the Γ_7 symmetry but $CeAgSb_2$ is in the Γ_6 symmetry. The mutually different LDs have also been seen for the cubic Pr compounds, PrB_6 in the Γ_5 symmetry, $PrIr_2Zn_{20}$ in the Γ_3 symmetry, and $PrBe_{13}$ in the Γ_1 symmetry [5,9].

As we have already shown for tetragonal $YbCu_2Si_2$ [2], the excited 4f-orbital symmetry can also be probed by the temperature dependence of LD originating the partial occupation of the excited states. We have applied this technique to cubic $PrBe_{13}$, where the LD is reduced at 50 K compared with that at 10 K without changing its sign. From our analysis, it is found that the first-excited states are in the Γ_4 symmetry with ~70 K for $PrBe_{13}$ [11].

Our work has been performed under collaboration with H. Hidaka, T. Yanagisawa, H. Amitsuka (Hokkaido U), T. Onimaru, T. Takabatake (Hiroshima U), E. Ebihara (Shizuoka U), Y. Onuki (U Ryukyus), S. Nakatsuji (ISSP, U Tokyo), T. D. Matsuda, Y. Aoki (Tokyo Metropolitan U), A. Kikkawa, Y. Taguchi (RIKEN) and F. Iga (Ibaraki U).

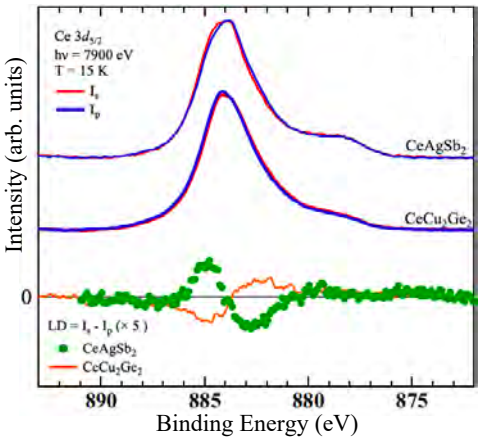

Fig. 1 (Upper) Linearly polarized angle-resolved Ce $3d_{5/2}$ core-level photoemission spectra of tetragonal $CeAgSb_2$ and $CeCu_2Ge_2$ along the [001] direction. (Lower) Comparison of the linear dichroism (LD) defined by the spectral difference between the s- and p-polarization configurations [7,10].

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Observation of low-energy fluctuations in $RE B_6$ (RE = Ce and Sm)

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Cubic rare earth hexaboride CeB_6 and SmB_6 have been studied by using nuclear quadrupole resonance (NQR) and low magnetic field nuclear magnetic resonance (NMR) techniques. CeB_6 is a prototypical compound for antiferroquadrupole (AFQ) ordering. A promising AFQ ordered structure at magnetic fields in this compound is so called O_{xy} type, which is evidenced mainly by the neutron scattering [1], NMR [2], and theoretical works [3]. In contrast, the ordered state in zero field has not been clarified to date due to lack of experimental information.

We have for the first time observed the ^{11}B-NQR signal of CeB_6 and found that the NQR spectrum does not show a change expected for the O_{xy} type order within experimental accuracy. For further examining the state at very low fields in CeB_6, ^{11}B-NMR measurements were performed at fields below 0.1 T. When the system is in the AFQ ordered state and in field, secondarily induced internal field brings about changes in the NMR spectral shape. It is indeed clearly observed above 0.1 T, while it is less emphasized with decreasing external field.

The obtained powder pattern spectrum is well reproduced by considering the field-angle dependence of the resonance lines of a single crystalline sample reported by Takigawa *et al* [4]. The field-induced internal field estimated by analyzing the present spectral data decreases with lowering external field and was found to be extrapolated to zero at a finite field of about 0.06 T. Thus these results of ^{11}B-NQR and ^{11}B-NMR measurements suggest that the AFQ ordered phase in zero field differs from that above 0.1 T.

We have also succeeded in observing the ^{11}B-NQR signal of isostructural compound SmB_6. SmB_6 has been known as a typical compound of Kondo insulator as well as valence fluctuation. Recently its interest has been renewed, since a new viewpoint of a topological insulator was proposed [5]. An early study of ^{11}B-NMR measurement indicates the existence of low-energy magnetic fluctuations in this compound [6]. Although the origin of the low-energy magnetic fluctuations, especially whether or not there is any relationship between the magnetic fluctuations and the surface properties, is a problem to be solved urgently. However experimental information at zero and very low magnetic fields is lacking, and therefore it is expected that our measurements at zero and very low fields will bring crucial information about that.

We will present our new results and progressed analyses of SmB_6 as well as CeB_6.

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Theoretical Aspects of Valence Fluctuation Systems Emerging in Sm and Eu Compounds

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Through the J-Physics project, we have mainly studied the aspects of valence fluctuation and Kondo phenomena in Sm and Eu compounds [1, 2]. Today, I would like to report a part of those attempts in this presentation. It is well known that the valence fluctuation takes place between f^5 and f^6 configurations in the Sm systems and between f^6 and f^7 configurations in the Eu systems. The common key ingredient to understand the phenomena is the many-body singlet ground state and the low-lying excitations in the f^6 configuration.

The first topic is the field-insensitive heavy-fermion (HF) states found for the Sm skutterudite and the Sm 1-2-20 compounds. We introduced a two-orbital impurity Anderson model to describe a competition of the f^6 local singlet with an orbital Kondo singlet in f^5 configuration. The model was shown to exhibit a quantum critical point (QCP) between the two configurations. Then we studied the Fermi-liqud properties of the model around the QCP via the numerical renormalization group method. It was clearly found that a field-insensitive Fermi-liquid state with a significantly enhanced γ value of the specific heat emerges in the vicinity of the QCP. The relevance of these results to the real materials are discussed.

In the case of Eu , the f^7 configuration gives rise to the stable large-spin state with S=7/2, which normally results in a magnetic long-range order in the absence of the Kondo effect. Then, it is interesting to explore how the order is stabilized from the f^6 singlet ground state through the variation of the f-electron occupancy. To answer this question, we introduced an effective periodic Anderson model that consists of two distinct types of f electrons with itinerant and localized characters, in which six f electrons are assumed to be localized and to form the J multiplet structure. We analyzed the mean-field solution of the model and investigated particularly the stability of the ferromagnetic (FM) state under an interplay between the Hund's rule coupling and c-f hybridization strength (Fig. 1). Making use of the boson expansion for the localized f-electron operators, we also studied the magnetic excitation in the vicinity of the transition (Fig. 2). It was found that significant softening and broadening of the excitation spectra are induced by valence fluctuation, suggesting the importance of the local excited states in the mixed-valent region.

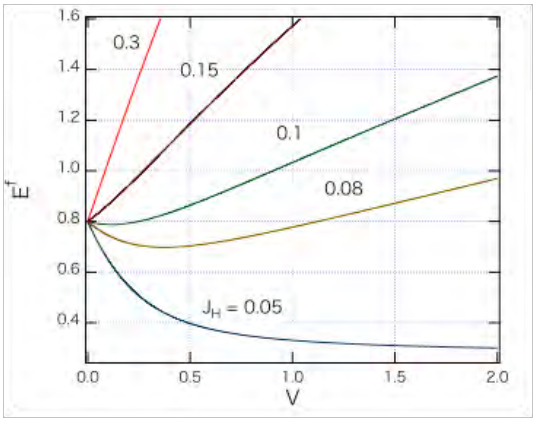

Fig. 1. Phase boundary of the Eu model between the PM and FM states for f-level energy Ef vs. c-f hybridizarion V; J_H represents Hund's coupling.

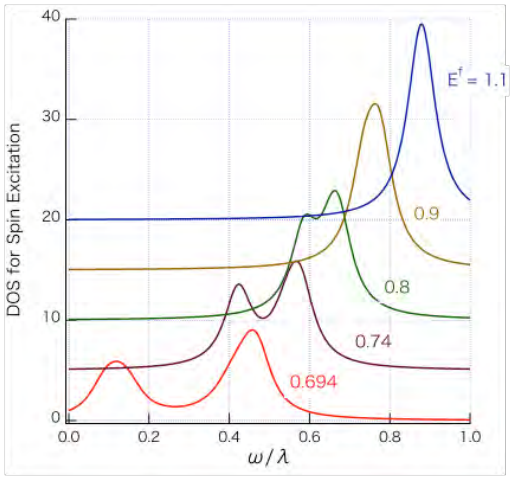

Fig. 2. Density of states of the magnetic excitation J = 0 → 1 in the PM states of the Eu model.

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Spin Nematic Liquids of Low-Dimensional Quantum Spin Systems

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The spin nematic phase, which is a kind of multipole phases, has attracted a lot of interest in the field of the strongly correlated electron systems, as well as the quantum spin liquid phase. Using the numerical exact diagonalization, the density matrix renormalization group (DMRG) calculation, and the finite-size scaling analysis, it is found that some spin nematic and spin liquid phases are induced by external magnetic field in the anisotropic and/or frustrated quantum spin systems. In our previous work[1], it was found that a field-induced nematic phase appears at some critical field in the anisotropic spin ladder and the mixed spin chain. The nematic phase is characterized by the power-law decay in the correlation function of the second-order spin moment. In addition at some higher critical field a quantum phase transition can occur to the conventional field-induced Tomonaga-Luttinger liquid. Several typical magnetization curves calculated by DMRG are presented.

Recently the field-induced nematic phase was observed on the frustrated spin ladder system[2]. So we study on a frustrated spin ladder system[3], using the numerical diagonalization and DMRG. As a result, it is found that several exotic quantum phases, including the spin-nematic liquid phase. We also report some exact eigenstates of the present model and present several interesting phase diagrams[4].

Our recent numerical diagonalization study[5] on the S=1 spin ladder system with the easy-axis single-ion anisotropy suggested that the field-induced nematic Tomonag-Luttinger liquid (TLL) phase appears. In addition another spin nematic liquid phases were predicted in the three-leg ladder system[6] and the three-leg tube system[7] with the ring exchange interaction. Some realistic candidates which possibly exhibit the spin nematic phase will be proposed.

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Kondo effect and magnetism in non-centrosymmetric f-electron materials

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The combination of strong spin-orbit interaction (SOI) and strong correlations has recently aroused great interest. In heavy elements, SOI can become large and strongly affects low-temperature properties resulting in fascinating phenomena such as the Quantum Spin Hall effect or the magnetoelectric effect. On the other hand, in materials with partially filled f-electron orbitals, strong electron correlations can commonly be observed, which results in intriguing physics such as magnetism, quantum criticality, and unconventional superconductivity. The combination of both, strong correlations and strong spin-orbit interaction, can thus be expected to result in entirely novel phenomena such as spin-orbit assisted Mott insulators, spin liquids, correlated topological insulators.

In this presentation, I will analyze the impact of the Rashba interaction in non-centrosymmetric systems on two phenomena commonly observed in f-electron materials, namely the Kondo effect and magnetism. In the first part, I will explain the emergence of so-called exceptional points and Fermi arcs in the single-particle spectral function at the Kondo temperature originating in non-hermitian properties of the Green's function matrix.

In the second part, I will analyze magnetic phases occurring in non-centrosymmetric f-electron materials. I will show the existence of a metallic antiferromagnetically ordered phase, which arises due to the interplay between Rashba interaction and a local hybridization. I will also analyze the transport properties of this phase, which are potentially useful for spintronics applications.

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Charge transfer effect in odd-parity crystalline electric field and critical volume effect

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Unconventional quantum criticality observed in the heavy-electron metal β-$YbAlB_4$ has attracted great interest [1]. The criticality is common to those discovered in the quasicrystal (QC) $Yb_{15}Al_{34}Au_{51}$ [2], which is well explained by the theory of critical Yb-valence fluctuations (CVF) [3]. Recently, direct evidence of the quantum valence criticality has been observed in α-$YbAl_{1-x}Fe_xB_4$ (x=0.014) [4] as well as QC $Yb_{15}(Al_{1-x}Ga_x)_{34}(Au_{1-y}Cu_y)_{51}$ (x=0, y=0) [5].

The CVF are charge-transfer fluctuations between the 4f and conduction electrons. To clarify the general property of the charge-transfer effect under odd-parity crystalline electronic field (CEF), we studied the electronic state in β-$YbAlB_4$ theoretically [6]. By analysing the CEF on the basis of the hybridization picture, odd-parity CEF is shown to exist because of sevenfold configuration of B atoms around Yb, which breaks the local inversion symmetry at the Yb site. This allows onsite admixture of 4f and 5d wave functions with a pure imaginary coefficient at Yb, giving rise to the magnetic toroidal (MT) degree of freedom. By constructing a realistic minimal model for β-$YbAlB_4$, we show that onsite 4f-5d Coulomb repulsion drives charge transfer between the 4f and 5d states at Yb, which causes divergently enhancement of the MT-dipole fluctuation as well as the electric-dipole fluctuation simultaneously with the CVF at the quantum critical point of the valence transition [6].

Recently, *non*-divergent Grüneisen parameter Γ toward the lowest temperature T=70 mK has been observed in the QC $Yb_{15}Al_{34}Au_{51}$ [7]. Surprisingly, its absolute value |Γ| at T=70 mK is smaller than that in the approximant crystal (AC) $Yb_{14}Al_{35}Au_{51}$, which shows the Fermi-liquid behavior. This poses a serious challenge to the conventional understanding that |Γ| diverges at *any* QCPs [8].

To clarify the mechanism, first we constructed the complete framework for calculating the specific heat C, thermal-expansion coefficient α, and Grüneisen parameter Γ near the magnetic QCP on the basis of the theory of spin fluctuations [9,10]. Then, we have applied this formalism to the case of the CVF near the QCP of the valence transition. We have shown that the measured behaviors of C, α, and Γ in the QC $Yb_{15}Al_{34}Au_{51}$ are naturally explained, which are consistent with the robust criticality under pressure [11]. The difference in Γ at the lowest temperature between the QC and AC is shown to reflect the difference in the pressure derivative of characteristic energy scales of the CVF and Kondo temperature [11].

In the presentation, we discuss these newly clarified aspects of the quantum critical phenomena in strongly-correlated electron systems on periodic and aperiodic crystals.

This presentation is based on the work done in collaboration with K. Miyake.

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New formula for nonlocal interactions based on dynamical mean-field theory and its applications

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Theoretical treatment of multipolar ordering often relies on effective local models such as Heisenberg model and RKKY-interaction model. These effective models are derived by considering strong-coupling limit of electronic models and projecting out the charge degrees of freedom. For descriptions of individual materials, one employs electronic structure calculations and evaluate effective nonlocal interactions J_{ij} in each material. In this way, we can address magnetic and multipole properties of individual compounds and even make a prediction of symmetry breaking in experimentally unknown materials.

Magnetic and multipolar ordering typically occurs in system with strong interactions, where theoretical approach using effective local models is justified. However, first-principles calculation is not justified in strongly correlated systems and often fails in obtaining a correct electronic structure. This motivates us to employ the dynamical mean-field theory (DMFT) and take strong correlations into account in the single-particle excitations. This approach is called LDA+DMFT or DFT+DMFT and have elucidated single-particles properties in strongly correlated materials.

We have derived a new formula for nonlocal interactions within DMFT. In the strong-coupling limit, this formula reproduces the kinetic exchange interactions in Hubbard model and the RKKY interactions in the periodic Anderson model. Furthermore, this formula is applicable away from strong-coupling limit, where local moments are not well defined anymore. Therefore, this new formula allows systematic investigations of nonlocal interactions in realistic strongly correlated materials. In this talk, some demonstrative results and applications to materials will be presented.

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NQR study of Ce_3PtIn_{11}

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^{115}In Nuclear Quadrupole Resonance (NQR) measurements have been carried out on Ce_3PtIn_{11} [1], a heavy fermion compound harboring two inequivalent Ce sites. This system exhibits magnetic transitions at T_{1n} = 2.2 K and T_N= 2 K, prior to entering a superconducting state below T_c= 0.32 K. Previous measurements imply that magnetism and superconductivity are spatially separated and only the Ce(2) sublattice orders magnetically, with a quantum critical point at pressure p_c = 1.3 GPa, while the Ce(1) sublattice is paramagnetic. However, the present nuclear relaxation rate behavior of $1/T_1 \sim T^{0.5}$ reveals the presence of critical spin fluctuations indicative of close proximity to another quantum critical point, presumably at a small negative pressure, which is associated solely with the Ce(1) sublattice. The ordered wave vector is found to be most likely *(1/2, 1/2, h)*.

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J-Physics Annual Meeting FY2019, January 6–8, 2020, Centennial Hall, Kobe University

Study of Crystalline-Electric-Field States in Yb-diluted Systems $Y_{1-x}Yb_xT_2Zn_{20}$ (T = Co, Rh)

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The YbT_2Zn_{20} family (T = Cobalt-group elements) is known as an Yb-based heavy fermion system [1-4]. As shown in Fig. 1, the crystalline-electric-field (CEF) states of these compounds were estimated from the results of macroscopic measurements. One of the unsolved problems in this system is an unusual evolution of the CEF level scheme from the Co system (Γ_6- Γ_8- Γ_7) to the Rh and Ir systems (Γ_6- Γ_7-Γ_8), which is difficult to explain with a conventional point charge model described by Lea et al [5]. In a conventional model, a sign change of the CEF parameter (B_{60}) may be needed to explain such evolution of CEF states, indicating the existence of a structural transformation and/or a change of the coordination charge on ligands. However, no evidence of a drastic change on the ligands has been reported. To solve this problem, we have started to examine the CEF states of the Co-group compounds by means of X-ray diffraction (single-crystal structure analysis), neutron scattering, and single-ion magnetic anisotropy measurements. In this presentation, we show results of magnetization measurements for (Y, Yb)Co_2Zn_{20} and (Y, Yb)Rh_2Zn_{20} (not shown in this abstract).

Magnetization measurements were carried out by utilizing a commercial SQUID magnetometer (Quantum Design, MPMS) for H || [100], [110], [111] with single-crystalline samples of the Yb-diluted systems $Y_{1-x}Yb_xT_2Zn_{20}$ (T = Co, Rh). In Fig. 2, we show the field dependence of magnetization; here, the magnetization data (open circles) were normalized, and the background components were subtracted so that the data of $YbCo_2Zn_{20}$ below 2 T are roughly reproduced with those of the diluted system (a careful analysis for high-field data is in progress). As shown in this figure, the magnetic anisotropy of the diluted system is qualitatively the same as that of the parent compound (solid lines). In addition, we confirmed that the evaluated CEF parameters for the diluted system is in good agreement with those of the $YbCo_2Zn_{20}$ system.

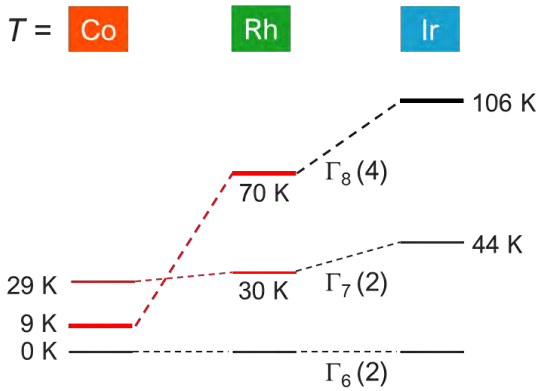

Fig. 1. A proposed CEF level scheme of YbT_2Zn_{20} [2-4].

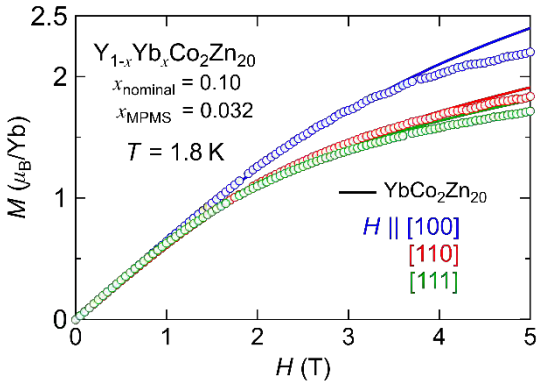

Fig. 2. Magnetic field dependence of magnetization of $Y_{1-x}Yb_xCo_2Zn_{20}$. The composition of Yb, x_{MPMS}, was estimated from the magnetization data.

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Probing Multipolar Magnetism in Pyrochlore Materials with Magnetostriction

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Multipolar magnetism is an emerging field of quantum materials research. The building blocks of multipolar phenomena are magnetic ions with a non-Kramers doublet, where the orbital and spin degrees of freedom is inextricably intertwined, leading to unusual spin-orbit entangled states. The detection of such subtle forms of matter has, however, been difficult due to a limited number of experimental tools.

Quantum spin liquids (QSLs) and Multipolar ordered states (MPOs) both share the property of being notoriously difficult to detect with conventional probes. The pyrochlore oxide family offers a unique arena to investigate the collaboration between QSLs and MPOs. For instance, $Pr_2Zr_2O_7$ is known to host a non-Kramers doublet in the ground state where time-reversal even electric quadrupolar moments are active. These moments reside on a pyrochlore lattice, where frustrated pairwise interactions allow the possible existence of QSL with an emergent U(1) gauge field and accompanying bosonic spinons, known as quantum spin ice.

Recently, lattice-based techniques are proposed as novel probes to detect MPOs [1]. Motivated by this success, we investigate the possibility of detecting QSL when it arises from interacting multipolar moments. In this poster presentation, we theoretically propose that spin-lattice coupling can be used as a powerful tool to probe such multipolar QSL phases.

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Classification of superconducting pairing induced by fluctuation of momentum-based multipoles

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Recent studies of superconductivity have focused on spin fluctuation, instead of electron-phonon coupling, as an origin of attractive interaction between electrons. Such superconductivity mediated by fluctuations of electric or magnetic orders is a central issue in this field.

On the other hand, a multipole order, which represents electrons' degrees of freedom in strongly spin-orbit coupled systems, is attracting much attention. Therefore, it is helpful for understanding unconventional superconductivity to consider "superconductivity induced by fluctuations of multipole orders," as an extension of spin-fluctuation-mediated superconductivity. Indeed, previous works have studied superconductivity induced by fluctuations of *odd-parity electric* multipole orders [1, 2]. Especially, Ref. [1] suggested that odd-parity (spin-triplet) superconductivity is realized by a ferroic fluctuation of odd-parity electric multipoles, using the multipole classification in *isotropic* systems [3].

Stimulated by the above backgrounds, we study superconductivity induced by fluctuations *odd-parity magnetic* multipoles, aiming at good understanding of multipole-fluctuation-mediated superconductivity. Furthermore, this study focuses on the effect of *crystalline electric fields* using the classification theory of multipoles [4, 5], while the previous studies [1, 3] simply assume isotropic systems. As a result, we suggest that a nodal extended *s*-wave pairing may be favored by an odd-parity magnetic multipole fluctuation under crystalline electric fields.

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NMR evidence of a non-magnetic phase transition in CeCoSi

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CeCoSi crystalizes in the tetragonal structure with a *P*4/*nmm* (D_{4h}^7, No. 129) space group without inversion symmetry at the Ce site. This system has two ordered phases: the antiferromagnetic (AFM) transition occurs at T_N = 9.4 K [1,2], and another transition is found at T_0 = 12 K above the AFM phase [3]. The AFM phase is abruptly suppressed by hydrostatic pressure at ~ 1.3 GPa, whereas T_0 gets maximum at ~ 1.5 GPa and reaches T_0 ~ 40 K, and then, it is suppressed at ~ 2.15 GPa [1,2]. Several interesting scenarios are proposed for this ordered phase including multipolar ordering or spin-density-wave ordering.

We performed ^{59}Co nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) measurements on a single-crystalline CeCoSi sample at ambient pressure and under pressure to reveal the origin of the anomaly below T_0. The NQR parameter ν_Q, which is proportional to the electric field gradient at the nuclear site, exhibits a kink at T_0, while no split nor broadening was detected in the NQR spectra without an external field. The nuclear spin-lattice relaxation rate $1/T_1$ exhibits little anomaly at T_0, suggesting the absence of the enhancement of the magnetic fluctuations. These results indicate a non-magnetic phase transition at T_0. On the other hand, the NMR spectra split below T_0 with the field along the [100] axis under pressure, as shown in Fig. 1. This result shows the symmetry reduction of the CeCoSi below T_0. We speculate that an antiferroquadrupolar (AFQ) ordering is a promising phase below T_0.

In my presentation, I will show the recent NMR and NQR results on CeCoSi including those in the AFM state. I will discuss the origin of the anomaly in the NMR and NQR, and discuss how these data are explained based on the scenario of the AFQ ordering.

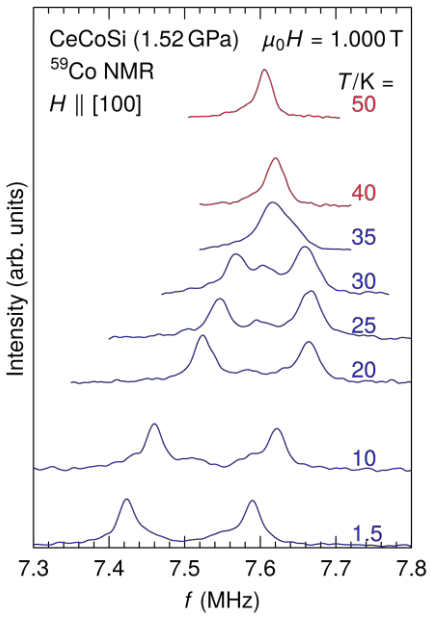

Fig. 1. Temperature dependence of the ^{59}Co NMR spectra of CeCoSi at 1.52 GPa with the field along the [100] direction. The spectra shown above are the third satellite line of the nuclear quadrupole splitting.

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Theory of Photon-Assisted Magnetoacoustic Resonance as a New Probe of Quadrupole Dynamics

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Motivated by the recent experimental development of detecting sideband nuclear magnetic resonance peaks assisted by electromechanical phonons [1], we theoretically investigate a possibility of hybrid measurement using electron paramagnetic resonance and a surface acoustic wave (SAW) [2]. Considering an extremely strong quadrupole-strain (QS) couplings suggested for silicon vacancies [3,4], we present a minimum model of the two-level system to study a magnetoacoustic resonance (MAR) coupled to various strain modes driven by the SAW. Using the Floquet theory [5,6], we elucidate the roles of longitudinal (A_L) and transverse (A_T) QS couplings in a photon-assisted single-phonon transition process, which are changed by rotating an applied magnetic field [2].

The left and middle panels in Fig. 1 show the time-averaged single-phonon transition probability for $\varepsilon_0 = \omega$ (the energy splitting equals the phonon frequency) and the QS coupling magnitudes normalized by ω, respectively, as a function of the field direction θ. For a cubic crystal field, there are two QS couplings g_3 and g_5 classified to the Γ_3 and Γ_5 point-group characters, respectively. The transition probability exhibits an abrupt decrease in $0 < \theta < \pi/8$ for the Γ_3-dominant coupling ($g_3 > g_5$) and in $\pi/8 < \theta < \pi/4$ for the Γ_5-dominant coupling ($g_3 < g_5$). This abrupt change in the θ-dependent transition probability is explained by the appearance of the sharp peak and the energy shift of the peak towards a lower value from $\varepsilon_0 = \omega$ in Fig. 1 (right). This is owing to the longitudinal A_L coupling effect assisted by a photon transition process, which becomes prominent at θ for the minimum $|A_T|$, and $|A_L|/\omega$ approaches 1.8 [2]. When θ differs from this angle, the peak broadens owing to the transverse A_T coupling effect. It is essential to find the resonance angle for evaluating the QS coupling ratio.

Thus, the MAR measurement proposed here is useful to confirm the existence of quadrupole degrees of freedom strongly coupled to elastic strains as observed in the silicon vacancy. In addition, it will also be applicable to phonon-mediated control of quantum spin devices.

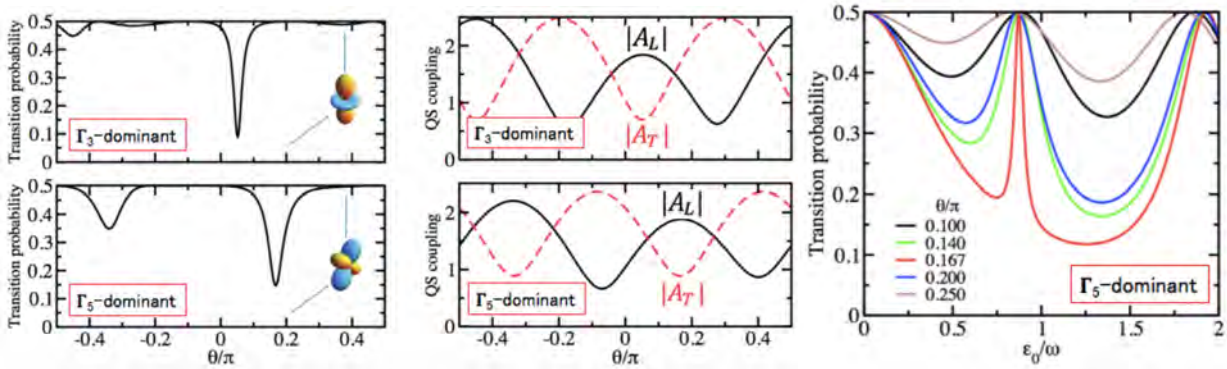

Fig. 1. (Left) Transition probability and (Middle) QS couplings plotted as a function of the field direction θ in the Γ_3-dominant and Γ_5-dominant cases. (Right) Transition probability plotted as a function of the energy difference ε_0 between the two states. The data are shown around $\theta/\pi = 0.167$ for the sharp peak in the Γ_5-dominant case.

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The study of Zn- and Fe-doping effect in T* type cuprates $La_{1-x/2}Eu_{1-x/2}Sr_xCuO_4$

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Recently, the local structure of high-T_c cuprates, in particular the oxygen around the cupper sites, are much attracted. These cuprates have three types of "214" structures; T-, T'-, and T*-types. In T'-type structure, in which a cupper atom is surrounded six oxygen atoms, the physical properties such as magnetism and superconductivity often change after annealed but the origin is unknown yet. The one of the possibility is the change of local structure associated with the oxygen sites [1]. On the other hand, the case of T*-type structure, in which a cupper atom is surrounded the oxygen atoms like the pyramid, has not been reported yet. In theses days, we are success to synthesize the T*-type cuprate $La_{1-x/2}Eu_{1-x/2}Sr_xCuO_4$ (LESCO) and search for the physical properties by magnetization, electrical resistivity, NMR, and μSR measurements [2]. We found that the mobility of hole with anti-ferro spin fluctuation is the important role of formation for the cooper pair but the superconducting properties is unknown yet.

To reveal the superconducting properties, we focus on the impurity effect. Zn and Fe are good elements as non-magnetic and magnetic impurities. In this presentation, the results of magnetization and μSR measurements will be reported and we are planning to discuss the superconducting properties in LESCO.

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Existence of ferromagnetic quantum critical point in Kondo lattice system $CeRh_6Ge_4$ investigated from microscopic and macroscopic measurements

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$CeRh_6Ge_4$ is a stoichiometric Kondo lattice with weak ferromagnetism below T_c = 2.5 K. It crystalizes in non-centrosymmetric hexagonal structure with the space group of *P-6m2*. The easy axis is perpendicular to the *c* axis, and the spontaneous magnetization of ~0.16 μ_B/Ce is also induced perpendicular to the *c* axis below T_c. The reduced ordered moment and the small entropy of ~20% of *R*ln2 just above T_c indicate the strong Kondo effect of this material. The relatively large electronic specific-heat coefficient (250 mJ/mol·K^2) implies that it would be in the vicinity of a ferromagnetic quantum critical point (FM QCP)[1]. However, in most clean metallic ferromagnets, a QCP at zero field is avoided [2]. For example, in CeRuPO, a QCP is avoided by changing the FM transition to antiferromagnetic transition [3]. Meanwhile, in UGe_2, a QCP is avoided by changing to discontinuous transition [4]. We measured electrical resistivity under pressure for $CeRh_6Ge_4$, and found that the Curie temperature of 2.5 K decreases continuously as increasing pressure and the FM state is easily suppressed by low pressure of P_C ~ 0.85 GPa without any clear signature that transition changes to first order [5]. In addition to this result, we found NFL behavior around P_C. These results suggest that there is a FM QCP at zero field or at least in the immediate vicinity of zero field in $CeRh_6Ge_4$. Recently, we checked the magnetic transition in $CeRh_6Ge_4$ at ambient pressure using NMR measurement. From microscopic measurement results, the transition is understood to be of second order. In the presentation, we will report both macroscopic and microscopic results.

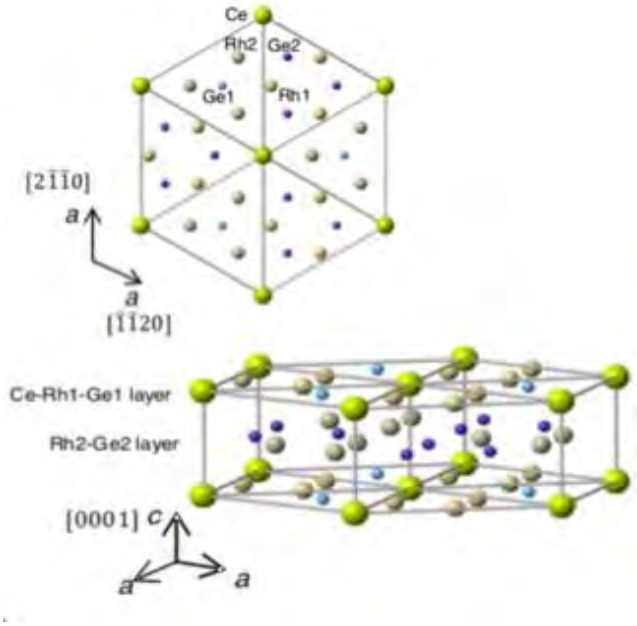

Fig. 1 The structure of $CeRh_6Ge_4$

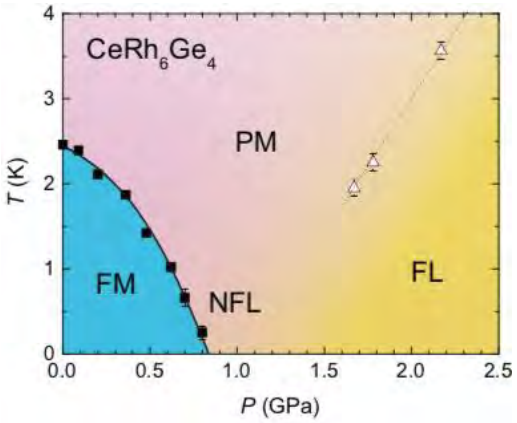

Fig. 2 The *P-T* phase diagram of $CeRh_6Ge_4$

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Odd-parity topological superconductivity in UTe_2

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Recent discovery of superconductivity in uranium ditelluride UTe_2 [1, 2] is attracting great interest. UTe_2 shows a superconducting transition temperature T_c~1.6 K and a nonmagnetic behaviour down to 25 mK [3]. A large upper critical field and re-entrant superconductivity have been observed by high-field experiments [1, 4, 5]. The Knight shift and $1/T_1T$ by nuclear magnetic resonance (NMR) experiments [6] are enhanced with decreasing temperature, indicating the enhanced magnetic fluctuations along *a* axis. It suggests the spin-triplet pairing induced by ferromagnetic fluctuation. Thus, UTe_2 is a strong candidate of odd-parity spin-triplet superconductors.

Identifying the topological nature has been one of the central issues in condensed matter theory. Because odd-parity superconductors are a strong candidate of topological superconductors, identifying topological properties of a fresh and good platform UTe_2 is desired. A nonmagnetic behavior of UTe_2 and a relatively high transition temperature at ambient pressure enables us to use many experimental tools. Theoretically, clarifying the Fermi surface is crucially important to detect the topological superconductivity.

Theoretical studies on superconductivity for this material suffered difficulty because first-principles calculations predict an insulating electronic state, incompatible with transport measurements as well as a recent ARPES measurement [7, 8]. This fact implies that the Coulomb interaction is crucially important for UTe_2.

In this work [9], we theoretically study the electronic state and superconductivity of UTe_2 by a GGA+*U* method. The GGA+*U* results shows an insulator-metal transition by Coulomb interaction for *U*~1.0 eV. The insulator-metal transition is a peculiar property of UTe_2, which was not observed in other uranium-based superconductors nor in Kondo insulators. Fermi surface formulas for the three-dimensional winding number and three two-dimensional Z_2 numbers indicate topological superconductivity for the intermediate *U*, and accompanied surface states are provided. We also predict the superconducting gap node at zero magnetic field and under magnetic field along *b* axis. Based on these results, we propose multiple superconducting phases under the magnetic field [4].

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Theoretical study of dichromatic transport of Mn-based odd-parity magnetic multipole system

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The multipole degree of freedom, which arises from the entanglement of spin, orbital, and sublattice, has attracted much attention. Particularly, peculiar itinerant properties and cross-correlated responses are induced by the spontaneous multipole order accompanying the parity violation, which is called as *odd-parity multipole order* [1]. Although there are a lot of theoretical/experimental efforts, further investigations should be required to elucidate the nature of odd-parity multipole order.

One of powerful probes of parity-violation is a *non-linear response*. For instance, the second order non-linear electronic conductivity (dichromatic transport/ non-reciprocal transport) is a characteristic response of parity-violated systems. Historically, the phenomenon was elaborately studied by Rikken et al., and it was revealed that the response is greatly enhanced in spin-orbit coupled semiconductor /superconductor [2]. Furthermore, a geometrical effect on non-linear responses has been extensively studied both in theoretical and in experimental studies [3].

In our work, we conduct theoretical study of dichromatic transport in the odd-parity magnetic multipole system. The system shows the parity-violation due to the magnetic order and possesses the itinerant property which is different from the systems in previous studies. We would discuss the symmetry analysis and calculations of the dichromatic transport with the minimal Hamiltonian modeling on Mn-based odd-parity magnetic multipole ordered materials [4].

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Spin-orbit-coupled metal candidate PbRe2O6

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Spin-orbit-coupled metals (SOCMs) have a specific Fermi liquid instability because of large spin orbit interactions [1]. In SOCM, the instability causes a phase transition from a symmetric to a space inversion symmetric phase and arises ferroelectric, gyrotropic and multipole phases. However, only two materials, $Cd_2Re_2O_7$ and $LiOsO_3$, are known for candidates of SOCM. Then, more model materials are highly desired for revealing the nature of instabilities in SOCMs.

In this study, we have investigated the lead rhenium oxide $PbRe_2O_6$ as the new candidate. The compound comprises a stack of modulated honeycomb lattices made of Re^{5+} ($5d^2$) ions in a centrosymmetric *R*–3*m* structure at room temperature [2]. As shown in Fig.1, Re_2O_{10} units, which is consisted of edge-shared ReO_6 octahedra, form a honeycomb network by sharing corners in the (0 0 1) plane. each Re-O layers are connected with sharing the corner of octahedra. There are two kinds of Pb atoms between the Re-O layers: the Pb1 atom is sandwiched by two triangular holes, which are inverted by inversion symmetry at the Pb1 atom, and the Pb2 is located between triangular and hexagonal holes. There is inversion symmetry only at Pb1 site. As shown in Fig.2, in our physical property measurements, two phase transitions are found at T_{s1} = 265 K and T_{s2} = 123 K. At T_{s1}, a structural transition to a monoclinic structure takes place and the density-of-states is enormously reduced, while small changes are observed at T_{s2}. Surprisingly. $PbRe_2O_6$ shares a lot of similarities with another SOCM candidate $Cd_2Re_2O_7$ in spite of the crucial differences in the crystal and probably in the electronic structure. The characteristics of $PbRe_2O_6$ will be presented, and the possibility of SOCM will be discussed by comparison with $Cd_2Re_2O_7$.

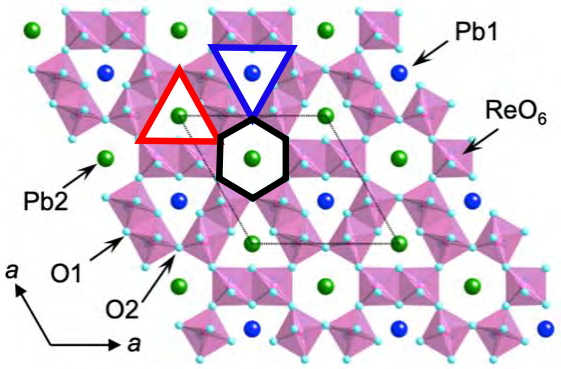

Fig. 1. Crystal structure of PbRe2O6

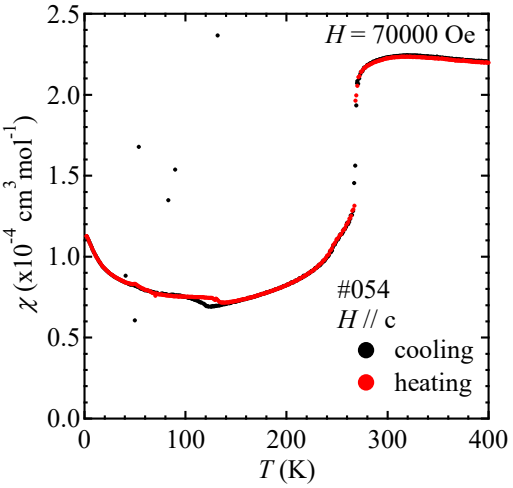

Fig.2. Temperature dependence of Magnetic susceptibility

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Structural Analyses and Superconducting Properties of New Caged Compounds

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Heavy fermion(HF) compounds with cage structure have attracted much attention due to a variety of interesting phenomena, caused by enhancement of *c-f* hybridization and small crystalline electric field splitting. Therefore caged compounds, such as filled skutterudite and RTr_2X_{20}(*R*: rare earth, *Tr*: transition elements, *X*: Al, Zn, and Cd) crystallized a cubic structure, are intensively investigated so far. Recently we succeeded in developing of new superconductors with caged crystal structure in Yb-based skutterudite and 1-2-20 systems, and some of La and Lu-based 3-4-13 and 5-6-18 systems.

Among the filled skutterudites, however, only three systems($YbFe_4Sb_{12}$ $YbFe_4P_{12}$, and $YbOs_4Sb_{12}$ have been reported[1,2], because of the difficulty of synthesizing crystals. It has appeared that the physical properties show a large sample dependence, which is mainly caused by the Yb-site filling fraction *x*. We tried to grow high quality single crystals of $YbOs_4Sb_{12}$ by a way of both of high pressure-synthesis and a flux method under ambient pressure. The composition and structural analyses were performed by the EDX and single crystal x-ray diffraction precisely. As a result, we succeeded in growing high quality single crystals. It was confirmed that the Yb-site filling fraction *x* is certainly 1 for the crystals grown under ambient pressure. The lattice parameter is obviously larger than that expected from $YbOs_4Sb_{12}$ of trivalent Yb. By using these single crystals, we performed the resistivity, specific heat, and magnetic measurements. At low temperatures, we have discovered superconducting state below ~ 0.4 K for the first time.

Recently, we have also discovered some Yb-, La-, and Lu-based superconductors with $CeCr_2Al_{20}$-type and a modified $Yb_3Rh_4Sn_{13}$-type structure[3,4]. These crystal structures have a characteristic cage structure formed by Al or Sn as similar as the filled skutterudite. In this presentation, we would like to discuss about these crystal structure analyses results and superconducting properties.

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Anisotropic Magnetic-Field-Induced Phases in Cubic Chiral Antiferromagnet EuPtSi

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The skyrmion in magnetic materials is a spin texture with a characteristic size caused by the competing interactions and emerges as mesoscale particle-like topological excitations. EuPtSi is most likely the first rare-earth based intermetallic compound in which the skyrmion lattice is recognized. This compound has the LaIrSi-type cubic crystal structure with the space group $P2_13$ (No. 198) as in B20 compounds.[1] The divalent Eu ions form a trillium lattice, which is a three-dimensional network of corner-sharing triangles similar to the network of Mn atoms in MnSi. Franco *et al.* measured the magnetic susceptibility and specific heat of polycrystal samples and found that EuPtSi shows an antiferromagnetic ordering below $T_N = 4.1$ K.[2]

Kakihana *et al.* succeeded in growing single crystals of EuPtSi and found that the characteristic field-induced phase, which is reminiscent of the A-phase in MnSi, appears in magnetic fields along the [111] direction.[3] The observed distinct additional Hall resistivity ρ_H and pronounced magnetoresistance ρ in the field-induced phase (A-phase) strongly suggest the formation of the skyrmion lattice. In fact, recent neutron diffraction [4] and resonant X-ray scattering [5] experiments revealed the existence of the skyrmion lattice in the A-phase of EuPtSi. Recently, the magnetic phase diagram for H // [001] was studied, and the B-phase was found in addition to the A'-phase.[6] Here, we studied precisely the angular dependence of these field-induced phases in EuPtSi by the electrical resistivity measurements. Figure 1 shows the angular dependences of the resistivity anomalies for the magnetic field direction from [001] to [100], [100] to [010], and [010] to [001]. Here, the field-induced A'- and B-phase disappear at $\theta \sim 30°$ and appear again at $\theta \sim 80°$. When the magnetic field is tilted from [001] to [110] or [1-10] direction, the A'- and B-phase disappear at $\theta \sim 10°$ and 25°, respectively, and the A-phase appears in the field angle $40° < \theta < 75°$, as shown in Fig. 2. The field-induced phase was not observed for H // [110] ([1-10]) direction.

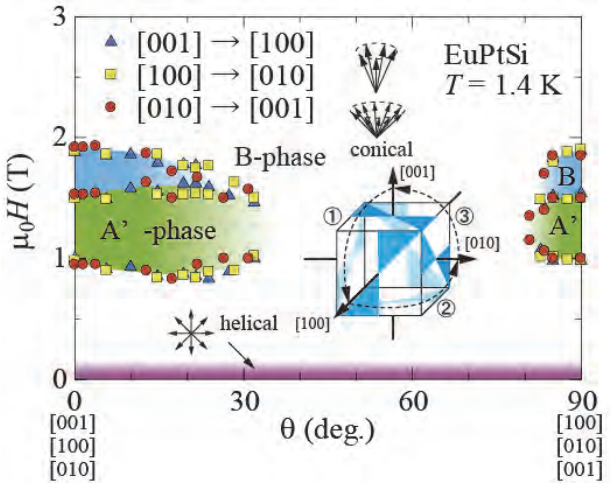

Fig. 1. Angular dependences of field-induced phases (A' and B-phase) from [001] to [100], [100] to [010], and [010] to [001].

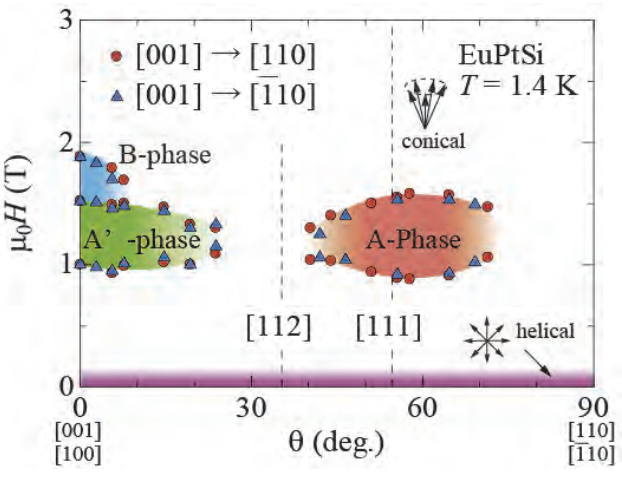

Fig. 2. Angular dependences of field-induced phases from [001] to [110], and [001] to [1-10].

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Chiral soliton lattice investigated by ac magnetoresistance in $Yb(Ni_{1-x}Cu_x)_3Al_9$

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Chirality plays an essential role in physical properties of materials with chiral crystal structures. Magnetic moments in chiral magnets take helical paths, due to Dzyaloshinskii-Moriya (DM) antisymmetric interaction. Recently, nonlinear magnetic order called chiral soliton lattice (CSL) has attracted much attention [1, 2]. The competition between DM interaction, Zeeman energy induced by a magnetic field, and symmetric exchange interaction such as Heisenberg exchange interaction and Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction results in the formation of macroscopic spin ordered structure such as CSL in chiral magnets. CSL appears in mono-axial chiral helimagnets such as hexagonal $CrNb_3S_6$ [2-4] and trigonal $YbNi_3Al_9$ [5-8], in which a linear magnetic order of a chiral helimagnetic structure is realized in zero magnetic field. When a magnetic field perpendicular to the chiral helical axis is applied, CSL is stabilized in these materials. One of the most impressive physical responses of CSL are discrete magnetoresistance (MR) [9] owing to the coupling between conduction itinerant spins and the localized spins with chiral magnetic order.

In $YbNi_3Al_9$ (space group: $R32$ #155, D_3^7), mono-axial chiral magnetic order with magnetic propagation vector $q = 0.8c^*$ is observed below 3.4 K in $YbNi_3Al_9$ [5-8], and CSL appears below a magnetic field of 0.1 T perpendicular to the chiral helical axis. In submillimeter sized $YbNi_3Al_9$, MR discretely jumps due to quantization of the soliton number in CSL [9].

In $Yb(Ni_{1-x}Cu_x)_3Al_9$, where Ni atoms are substituted with Cu atoms in $YbNi_3Al_9$, CSL is realized below the magnetic ordering temperature of 6.4 K and a critical magnetic field of 1 T, and these values become 2 and 10 times as high as those of $YbNi_3Al_9$, respectively [6]. Moreover, q is reduced to about $0.44c^*$ in $Yb(Ni_{0.94}Cu_{0.06})_3Al_9$ [10], indicating that carrier doping have effects on the magnetic interaction in $YbNi_3Al_9$.

In this work, we reinforce CSL state via element substitution effect in $Yb(Ni_{1-x}Cu_x)_3Al_9$, and measure AC MR in order to investigate CSL formation. One of the interesting points of this measurements is that a time-varying current may drive the soliton number of CSL via magnetoelectric effect which definitely arises in crystals with chiral spin ordered structure, because these crystals are noncentrosymmetric. We observe discrete MR with a small peak in each discrete point in $Yb(Ni_{0.94}Cu_{0.06})_3Al_9$ at 2 K. We will address the details of the results of the AC MR measurements of $Yb(Ni_{1-x}Cu_x)_3Al_9$.

This work is supported by Grants-in-Aid for Scientific Research on Innovative Areas "J-Physics" (Grants No. 18H04315) from MEXT, KAKENHI (18K03539, 17H02815), *Core-to-Core* Program “A Consortium to Exploit Spin Chirality in Advanced Materials” from JSPS, and the Naito Research Grant.

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Superconductivity in a new layered triangular-lattice system Li_2IrSi_2

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5*d* transition-metal compounds are expected to exhibit exotic phenomena such as pseudo-spin and mixed parity superconductivity, because their spin-orbit coupling (SOC) is significantly stronger than that of 3*d* transition-metal compounds. Several Ir-based superconductors have been reported, which include Li_2IrSi_3 [1,2]. However, to our knowledge, Li_2IrSi_3 (T_c = ~3.8 K) is the only alkali-metal compound reported till date. Therefore, the alkali-metal Ir-Si ternary system is an unexplored subject in new materials research, which motivated us to perform this study. In this presentation, we report the discovery of a new layered triangular lattice of Li_2IrSi_2 and the basic superconducting properties derived from magnetic susceptibility, electrical resistivity and heat capacity measurement [3].

It has a Ag_2NiO_2-type structure (space group *R*-3m) with the lattice parameters a = 4.02830(6) Å and c = 13.16180(15) Å (Fig.1-a). The crystal structure comprises $IrSi_2$ and double Li layers stacked alternately along the *c*-axis. The $IrSi_2$ layer includes a two-dimensional Ir equilateral-triangular lattice. Electrical resistivity and static magnetic measurements revealed that Li_2IrSi_2 is a type-II superconductor with critical temperature (T_c) of 3.3 K (Fig.1-b,c). We estimated the following superconducting parameters: lower critical field $H_{c1}(0) \sim 42$ Oe, upper critical field $H_{c2}(0) \sim 1.7$ kOe, penetration depth $\lambda_0 \sim 265$ nm, coherence length $\xi_0 \sim 44$ nm, and Ginzburg-Landau parameter $\kappa_{GL} \sim 6.02$. The specific-heat data suggested that superconductivity in Li_2IrSi_2 could be attributed to weak-coupling Cooper pairs.

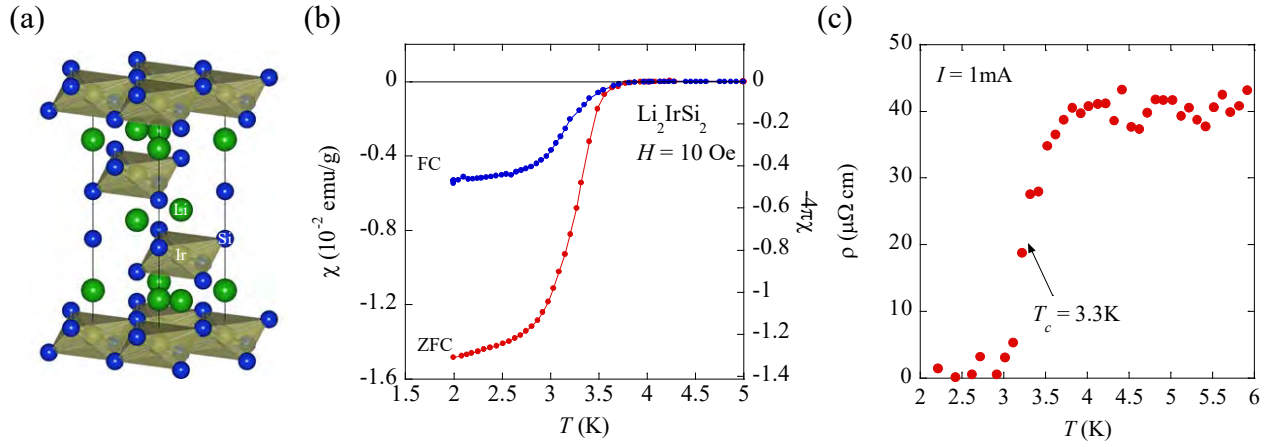

Fig.1 Crystal structure models for (a) Li_2IrSi_2 (space group: *R*-3m). (b) Temperature (T) dependence of the magnetic susceptibility (χ) of Li_2IrSi_2. (c) Temperature dependence of the electrical resistivity (ρ) of Li_2IrSi_2.

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Current-induced Novel Transport Properties in the Geometrically Frustrated Iridate $Ca_5Ir_3O_{12}$

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In recent years, there has been increasing interest in Ir oxides because of their attractive magnetic and transport properties due to strong spin-orbit interaction. $Ca_5Ir_3O_{12}$ has a hexagonal structure with noncentrosymmetric space group of $P\bar{6}2m$ (No.189). In the structure, 1D chains of the edge-sharing IrO_6 octahedra form triangular lattices in the *c*-plane. The average valence of Ir ions in $Ca_5Ir_3O_{12}$ is +4.67, so Ir^{4+} and Ir^{5+} exist in a ratio of 1:2. This situation can lead to the geometrical frustration of charge on both the triangular lattice in *c*-plane and 1D chains along the *c*-axis [1]. It is reported that $Ca_5Ir_3O_{12}$ exhibits an antiferromagnetic below 7.8 K and a second-order phase transition at 105 K [1]. The origin of this phase transition at 105 K is not clear at present; this is "hidden order" [1]. Recently, nonlinear electrical conductivity along the *c*-axis in a single crystal of $Ca_5Ir_3O_{12}$ was discovered in the non-ordered state [2].

To investigate the nonlinear electrical conductivity, we performed harmonic voltage response experiments. By using AC current, we can accurately measure the electrical conductivity when small amount of current is applied. Odd harmonics were observed below 200 K. The presence of harmonics means distortion of waveform, which is a proof of nonlinear conductivity [3]. Next, the resistivity of $Ca_5Ir_3O_{12}$ decreases with increasing the applied current because of its non-linearlity. Therefore, we have tried to carefully measure the change of thermoelectric power when current is applied. As a preliminary results, we have found that the additional thermoelectric power is induced with increasing current. We will report these results in this presentation.

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NMR study on high-field superconducting phase of Sr_2RuO_4

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Sr_2RuO_4, discovered in 1994[1], has been studied a lot because of its anomalous superconductivity though it is a very simple system. However, the superconducting symmetry is still unknown. Recent NMR study by A. Pustogow *et al*.[2], shows clear decrease of NMR Knight shift and this result give a strong constrain on the superconducting order parameter. We reproduce the decrease of NMR Knight shift[3].

The observed first-order superconducting-normal phase transition below 0.7 K indicates Pauli departing effect is dominant in Sr_2RuO_4. In addition, recent field-angle-dependent specific-heat measurements[4] implies that some anomalies exist in high-filed superconducting region.

In order to clarify the anomalies in low-temperature high-filed superconducting region of Sr_2RuO_4, we performed ^{17}O-NMR measurement under in-plane magnetic fields. To enhance the NMR signal intensity, we use ^{17}O substituted sample with T_c of about 1.5 K. Sr_2RuO_4 has two crystallographically non-equivalent oxygen sites, one at the RuO_2 plane and one at the apex. We mainly use the apical oxygen because of sharpness of the spectrum, and we observed splitting NMR spectrum in superconducting phase above 1.2 T, which is same region where the anomaly was detected by field-angle-dependent specific-heat measurement. In this poster presentation, I will show the preliminary experimental results of NMR measurements and discuss possible mechanism of NMR spectrum splitting and possible superconducting symmetry.

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^{125}Te-NMR study on the superconducting state of the spin-triplet superconductor UTe_2

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In the Uranium-based compounds of UGe_2, URhGe and UCoGe, spin-triplet superconductivity is strongly suggested to be realized on the basis of following experimental results[1]. (1) Superconducting (SC) phase overlaps or is inside the ferromagnetic (FM) phase in these compounds and (2) their upper critical field H_{c2} is far beyond the ordinary Pauli-limiting field. Furthermore, (3) it was shown that their superconductivity is strongly coupled with the Ising-type FM spin fluctuations, which are considered as a pairing interaction of the superconductivity.

In 2018, S. Ran *et al*. discovered a new U-based superconductor UTe_2 with SC transition temperature T_c = 1.6 K[2]. Although magnetic order is absent above T_c, different from other U-based FM superconductors, this compound also exhibits very large and anisotropic H_{c2}, exceeding the Pauli limiting field along the three crystalline principal axes[2, 3]. the nuclear spin-lattice relaxation rate $1/T_1$ in the normal state shows moderate Ising anisotropy of FM spin fluctuations[4]. These features suggest that UTe_2 is also a candidate of spin-triplet superconductivity. More interestingly, superconductivity in UTe_2 is enhanced when magnetic field along the *b* axis (the magnetic hard axis) is greater than 15 T[5, 6]. In addition, since the existence of a high-field SC phase exceeding 40 T[5] and multiple SC phases under pressure[7] have been reported, the SC properties of UTe_2 have attracted much interest and microscopic measurements in the SC state have been highly desired.

We performed NMR measurement on a single crystal of UTe_2 to investigate the SC properties[8]. The decrease of Knight shift in the SC state was much smaller than that estimated from Sommerfeld coefficient obtained from specific heat measurement (Fig. 1), indicating the spin part survives even in the SC state. In addition, $1/T_1$ does not show coherence peak just below T_c. These results are consistent with spin-triplet scenario. Furthermore, we found that the NMR linewidth in the SC state shows an unusual broadening.

We are measuring detailed NMR spectrum in the SC state to unveil the origin of the broadening of the spectrum. In this poster presentation, we will show abovementioned temperature dependence of $1/T_1$, Knight shift and linewidth of NMR spectrum in the SC state and discuss possible SC symmetry and the origin of the broadening of the spectrum.

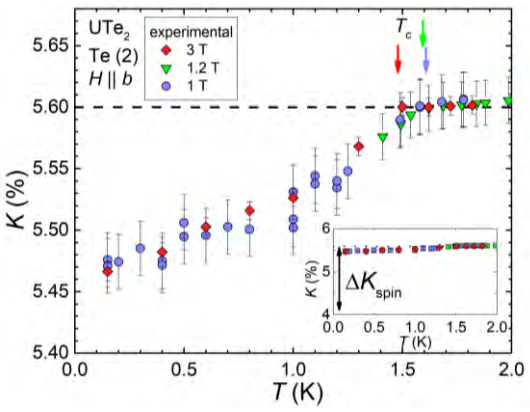

Fig. 1 Temperature dependence of the NMR Knight shift measured at several magnetic fields along *b* axis. The inset is the zoom-out view. The black double arrow in the inset shows the spin part of Knight shift estimated from the Sommerfeld coefficient.

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Possible Multipole Order without Local Inversion Symmetry in Tetragonal Antiferromagnet CeCoSi

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CeCoSi is a metallic antiferromagnet with T_N=9.4 K, and crystallizes into a tetragonal crystal structure ($P4/nmm$, D_{4h}^7), in which Ce atoms occupy the 2*c* cite ($4mm$, C_{4v}) [1]. Since the space inversion symmetry is locally broken at the Ce site, exotic phenomena originating from the so-called *odd*-parity multipoles may appear at low temperatures [2]. A unique pressure-temperature phase diagram of CeCoSi has been reported by Lengyel *et al* for the first time [3]. That is, (i) T_N remains nearly constant but suddenly disappears at P~1.3 GPa. (ii) A pressure-induced ordered phase (PIOP) with a super-zone gap formation abruptly appears just above P~1.3 GPa. (iii) The transition temperature of the PIOP reaches T_0~40 K at P~1.5 GPa. (iv) The PIOP is strongly suppressed with further applying pressures, and disappears at P_c~2.2 GPa. We investigated La-substitution effects and magnetization (M) under pressures in order to clarify the origin of the PIOP, and found that T_0 is suppressed by the La substitution and M shows not a cusp at T_0 but a further increase below T_0 as is frequently observed in compounds showing an antiferroquadrupole (AFQ) ordering [4]. Recently, we have succeeded in synthesizing CeCoSi single crystals and found a new transition at T_0~12 K, and concluded that an intermediate temperature range for $T_N<T<T_0$ is a part of the PIOP [5]. $P-T$ and $H-T$ phase diagrams are summarized in Figs. 1(a) and 1(b), respectively. In the workshop, we will present the bulk properties of $(Ce_xLa_{1-x})CoSi$.

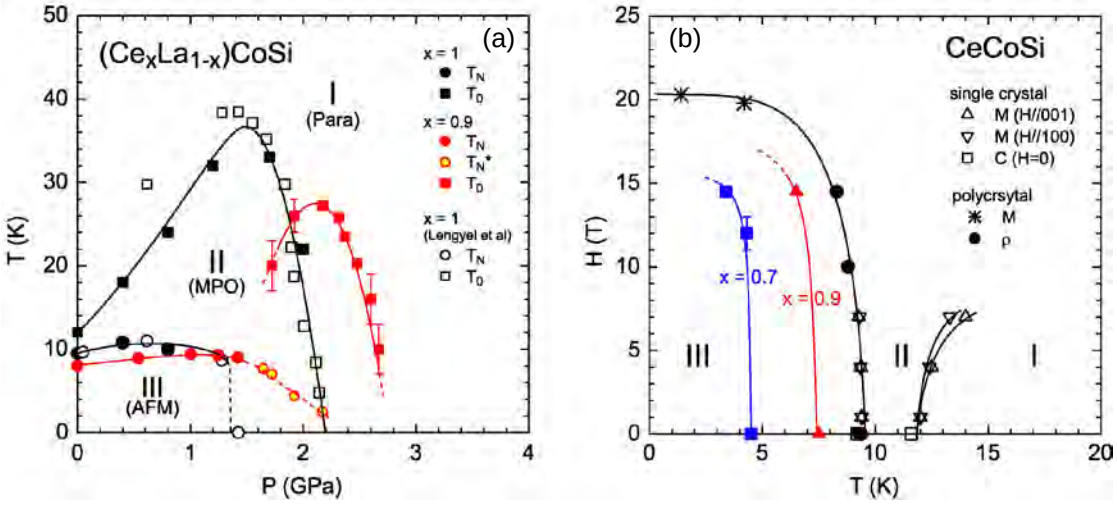

Fig. 1(a) Pressure-temperature phase diagram of $(Ce_xLa_{1-x})CoSi$ (x=1, and 0.9).
Fig. 1(b) Magnetic field- temperature phase diagram of $(Ce_xLa_{1-x})CoSi$ (x=1, 0.9, and 0.7)

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Thermal Expansion Measurement in $Cd_2Re_2O_7$

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The pyrochlore oxide $Cd_2Re_2O_7$ has attracted much attention as the first superconductor with T_c = 1.0 K in the family of pyrochlore oxide [1]. In addition to the superconductivity, the compound exhibits two successive phase transitions at T_{s1} ~ 203 K and T_{s2} ~ 112 K. Yamaura *et al*. suggested that these transitions are accompanied by characteristic structural changes from centrosymmetric cubic *Fd*-3*m* for phase I to noncentrosymmetric tetragonal *I*-42*m* for phase II and to another noncentrosymmetric tetragonal $I4_122$ for phase III [2]. On the basis of the suggested crystal structures for phase II and III, the successive transitions for $Cd_2Re_2O_7$ have been theoretically proposed to be orderings of odd-parity electric toroidal quadrupoles [3]; however, the space-group symmetry in phase II and III and their order parameters are still controversial. Interestingly, recent Cd-NMR measurements have indicated the possibility of an additional structural transition at T_{s1}' ~ 199 K (just below T_{s1}) [4]. In this case, the transitions at T_{s1} and T_{s1}' are considered to be structural ones from the cubic *Fd*-3*m* to cubic *F-43m* and to the tetragonal *I*-42*m*, respectively. On the other hand, no anomaly has been found at T_{s1}' in other bulk properties, such as electrical resistivity, specific heat, and magnetic susceptibility [1].

Thermal-expansion measurement will be quite useful to verify the presence of the unsettled phase transition at T_{s1}', since it generally can detect a tiny lattice distortion. In the present study, thermal expansion on single crystal of $Cd_2Re_2O_7$ was measured by the active-dummy method using two commercial strain gauges in the temperature range of 10-300 K. The measurements were made along the cubic [001] direction. Note that thermal expansion along the cubic [001] should be affected by uneven distribution of the tetragonal domains. Hereafter, linear thermal expansivity is denoted as $\Delta L/L_0$ ($\Delta L = L - L_0$ and L_0 refers to the value at 290 K).

The obtained temperature dependence of the linear thermal expansivity $\Delta L(T)/L_0$ above T_{s1} corresponds to the data calculated from the temperature dependence of lattice constant $a(T)$ in the previous X-ray diffraction measurements [5]. The present study reveals that $\Delta L(T)/L_0$ along the cubic [001] exhibits a clear kink anomaly at T_{s1}, but no anomaly at around T_{s1}' in the experimental accuracy. Similar results have also been reported in the thermal expansion measurements along the cubic [111] direction by Tachibana *et al* [6]. These facts do not support the presence of the cubic-tetragonal distortion at T_{s1}'. Below T_{s1}, our $\Delta L(T)/L_0$ data appear to correspond to the data calculated from $a(T)$ of the shorter tetragonal axis. At lower temperatures, $\Delta L(T)/L_0$ along the cubic [001] takes a minimum at T_{s2} = 111 K, while a much smaller anomaly can be found at T_{s2} in $\Delta L(T)/L_0$ along the cubic [111] direction [6]. The difference in the T_{s2} anomaly may be due to a difference in sample quality between the measured samples [1, 6].

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Study of Phonon Dispersion in the Geometrically Frustrated Iridate $Ca_5Ir_3O_{12}$ with Strong Spin-orbit Interaction

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Novel physical properties in iridium oxides is caused by strong spin- orbit interaction (SOI) and moderate electron correlation [1-3]. In the case of Ir^{4+} ($5d^5$), the strong SOI splits the six-fold degenerate t_{2g} states into the occupied $J_{eff} = 3/2$ and half occupied $J_{eff} = 1/2$ states. SOI changes a low energy band structure and Fermi surface. Inaddition, it is expected that SOI can also have important effects on phonon properties such as sound wave, specific heat, thermal conductivity, electron-phonon interaction, *etc.*

We reports the results of experiments and calculations of phonon dispersion of iridium oxide with a strong spin-orbit interaction (SOI) [4]. We measured the phonon dispersion in geometrically frustrated iridate $Ca_5Ir_3O_{12}$ by using inelastic X-ray scattering at room temperature (Fig. 1). We also show *ab inito* density-functional phonon dispersions based on the local density approximation and the generalized gradient approximation (GGA) considering SOI. By comparing experimental and calculated results, we found that the GGA phonon dispersion with SOI is in very good agreement with experimental result (See Fig. 1). The low-energy phonon properties such as the Debye temperature, specific heat, and sound velocity are also discussed.

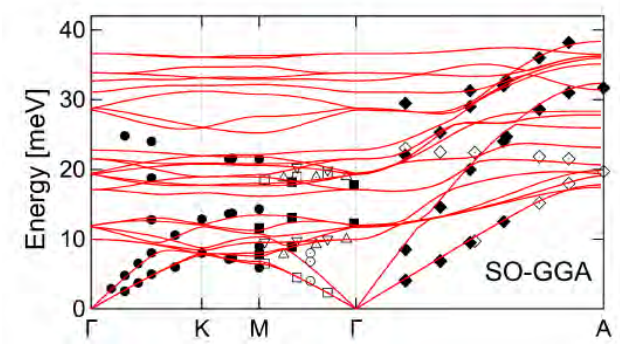

Fig. 1. Comparison with peak positions (symbols) and calculations obtained by GGA with SOI (SO-GGA) (red solid lines) in the reduced Brillouin zone [4].

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Raman scattering study of low-temperature structure and its symmetry on $Ca_5Ir_3O_{12}$

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$Ca_5Ir_3O_{12}$ has an intermediate valence Ir ion with the formal charge +4.67. From the valence state, we expect partially filled Jeff = 1/2 band, which is derived from strong spin-orbit interaction on Ir [1]. Although this leads to a metallic state, this compound shows insulative behaviour even at room temperature [2]. The origin of this insulative behaviour has not been revealed. The crystal structure is the regular triangular lattice of Ir-O chains, which are constructed by edge-sharing IrO_6 octahedra. Since this crystal structure has only one Ir site, a structural transition will be necessary to realize the insulative state. As $Ca_5Ir_3O_{12}$ shows a transition at 105 K, clarifying the structural change at this transition will give us a valuable insight to consider the electronic state on $Ca_5Ir_3O_{12}$.

We have measured Raman spectra on $Ca_5Ir_3O_{12}$ from 4 K to 300 K to elucidate the structural change [3]. The space group of the room temperature structure of $Ca_5Ir_3O_{12}$ is P-62m. The Raman scattering results have shown that the -6 symmetry conserves below 105 K, and that the O1 atoms will move in the *ab*-plane. In addition, the unit cell is expanded in the *ab*-plane. Since displacements in the *ab*-plane are important, we show the projected crystal structure along the *c*-axis in Fig. 1. The atomic positions are also described. As a candidate to explain the Raman scattering results, we propose the displacement of the O1 atoms as shown in Fig. 1 by arrows. This displacement pattern belongs to the irreducible representation A_2', and is a rotation around the -6 axis. To make a superstructure, it will be necessary to put the forward and reverse rotations around the -6 axes in an appropriate order. Though the size of the superstructure has not been clarified, we show an example of it with the super-lattice Bragg peak at the K-points, (1/3, 1/3, 0), in Fig. 2.

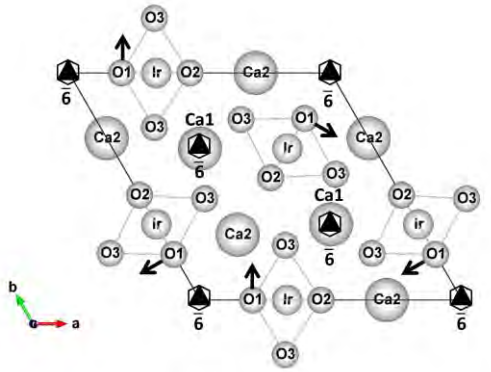

Fig. 1. The projected crystal structure along the c-axis. The label of atomic positions and -6 symmetries are shown. The arrows are the displacement of the A_2' mode of the O1 atoms.

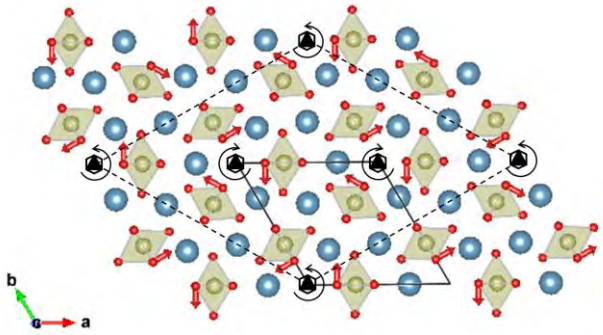

Fig. 2. An example of the superstructure. The -6 symmetries and the rotational direction around them are also displayed.

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Electric field gradient at Co site under antiferroquadrupole orderings in CeCoSi

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The concept of multipole is introduced for the multipole expansion in classical electromagnetics. It is used to represent electronic degrees of freedom in crystals, such as charge, spin, orbital, and their coupling. For example, anisotropic spatial distributions of electric charge and current for *f* electrons are denoted as the higher-rank multipole degrees of freedom, such as electric quadrupole and magnetic octupole, which result in multipole orderings [1]. Among them, the *f*-electron compound CeCoSi might be one of the potential candidates with such higher-rank multipole moments. The crystal structure is a centrosymmetric tetragonal structure (*P*4/*nmm*). The compound undergoes the antiferromagnetic (AFM) phase transition at T_N=9.4 K, whereas it shows another phase transition at T_0~12 K [2-4]. The latter phase, which is dominantly stabilized under pressure, is named as "pressure induced ordered phase (PIOP)". From the electrical resistivity measurement under the magnetic field, the PIOP phase is suggested to be an antiferroquadrupole (AFQ) ordered phase, although the order parameter in PIOP has not been identified yet. From the theoretical point of view, the AFM and AFQ states in CeCoSi would be interesting, since they are accompanied with the odd-parity multipoles if the staggered antiferroic multipole orderings which break the global inversion symmetry is realized [5-7]. From such an expectation, we focused on a possibility of odd-parity multipole ordered states in CeCoSi and have shown what type of odd-parity multipoles is possible [8]. Meanwhile, recent NMR and NQR measurements provide some information about the local electric field gradient at Co site [9]. As the lowering of the site symmetry at Co site largely depends on types of active multipole moments at Ce site, it is important to take into account these experimental findings.

In the present study, we study a possible electric field gradient at Co site in CeCoSi in the presence of the staggered AFQ orderings by concentrating on the PIOP phase. We investigate an effective localized model with the multipole-multipole interactions. By assuming the $\boldsymbol{q}$=**0** staggered inter-orbital AFQ ordered states with the moments Q_v (=Q_{x2-y2}), Q_{xy}, Q_{yz}, and Q_{zx}, we examine the NMR and NQR signals in each AFQ ordering. As a result, we find that the staggered AFQ ordering with the Q_v channel leads to additional signals in the NMR and NQR spectrum [10], which reflects the non-equivalent electric field gradient at Co site. On the other hand, the AFQ ordering with the Q_{xy}, Q_{yz}, and Q_{zx} channel does not change the number of the NQR signal, whereas NMR signal in those orderings depends on the direction of magnetic field. We discuss the obtained results in terms of the effective multipole couplings between magnetic dipole and electric quadrupole degrees of freedom, which will be useful to clarify the field-angle dependence of the NMR and NQR results.

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High-pressure NMR study of Dirac and Weyl semimetals, Black phosphorus and $MoTe_2$

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Two-dimensional layered Dirac and Weyl semimetals have attracted much attention due to their interesting physical properties. Of particular interest is the fact that external perturbations can change easily their physical properties. In Black phosphorus (BP), which is semiconductor at ambient pressure, band calculations predict that four twofold-degenerate Dirac corns appear at the Z point at P_c=1.2GPa [1]. In BP, the pressure is currently the most realistic external parameter to tune the band structure toward the formation of Dirac corns without introducing additional impurities, and therefore we have performed ^{31}P- nuclear magnetic resonance (NMR) measurements to investigate the band structure under P.

In order to investigate the band structure under pressure (P), we measured nuclear spin-lattice relaxation rate ($1/T_1$) which reflects the density of states (DOS) near the Fermi energy. In addition to the parameter of pressure, we have measured the field (H) dependence of $1/T_1$ under P. In our presentation, we present the details of P, T and H dependences of the $1/T_1$ and their implications in terms of the formation of the Dirac electrons.

In addition, we also have studied Type-II Weyl semimetal $MoTe_2$ [3], which shows a superconducting transition at 4K under pressure (2 GPa) [4]. In order to understand the superconductivity, we have employed the ^{125}Te-NMR technique under P (up to 2.17 GPa) and measured the NMR line profile and T_1, determining the Knight shift and low lying magnetic excitations. A superconducting signature has been observed in the temperature dependence both of K and $1/T_1T$ at around 2.5 K (2.17GPa, 0.46T). We present detailed NMR results and discuss a mechanism for the superconductivity from the microscopic point of view.

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Superconductivity in Mg_2Ir_3Si and Li_2IrSi_3: Fully Ordered Laves Phases

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The study of Laves phase, represented by the general formula AB_2, is a challenge in solid-state physics and chemistry for the following reasons. First, various structures such as *C*14, *C*15, and *C*36 (Strukturbericht symbols) appear because of the differences in the networks of the B_4 tetrahedra. It has not been fully understood which structure is obtained for given elements *A* and *B*. Secondly, a hidden Laves phase may emerge in a ternary system. It is referred to as a true ternary Laves phase when no binary Laves phase of its constituent elements exists. Hidden phases make the physics and chemistry of the Laves phase more diverging. Thirdly, some ternary systems exhibit fully-ordered phases in which two elements occupy B_4 tetrahedra in an ordered manner. The transition-metal elements (*TM*) and main group elements (*X*) are ordered as $TM_{3/4}X_{1/4}$ in the *B* site to form the Kagome (3636) network of the *TM* and the triangular (3^6) network of *X*. It has been unclear whether the ordered arrangement can be controlled.

Recently, we discovered a novel fully ordered Laves phase Mg_2Ir_3Si ($P6_3/mmc$, D_{6h}^4, No. 194) [1] and its superconductivity. A comparison of Mg_2Ir_3Si with another fully ordered Laves phase Li_2IrSi_3 ($P31c$, C_{3v}^4, No. 159) [2] demonstrated that the ordered arrangement of Ir and Si at the *B* site depends on the *A*-site elements. When *A* = Mg, an $Ir_{3/4}Si_{1/4}$-type ordering occurs and the breathing Kagome networks of Ir result [1]. In contrast, when *A* = Li, the sites occupied by Ir and Si are interchanged, and an $Ir_{1/4}Si_{3/4}$-type ordering emerges resulting in the breathing Kagome networks of Si, instead of Ir [2]. The change in the ordering pattern significantly influences superconducting properties: Mg_2Ir_3Si exhibits superconductivity at 7 K [1], which is markedly higher than the 3.8 K of Li_2IrSi_3 [2,3]. Our results will stimulate the further studies to determine the parameters that control the structures of the fully ordered Laves phases, and also suggest that the fully ordered Laves phase will provide a rich field for exploring new materials with interesting properties.

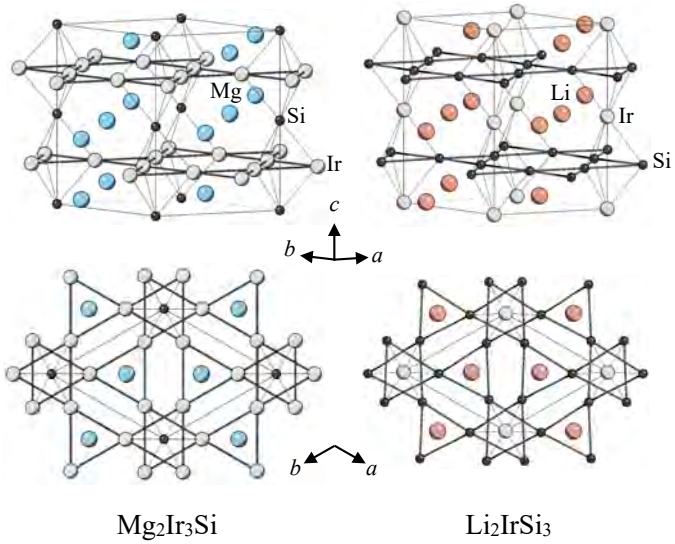

Fig. 1. Crystal structures of Mg_2Ir_3Si ($P6_3/mmc$, D_{6h}^4, No. 194) and Li_2IrSi_3 ($P31c$, C_{3v}^4, No. 159). These structures correspond to the fully ordered phase of $MgZn_2$ (*C*14). Solid lines indicate the unit cell.

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Magnetic responses and critical fields of the heavy-fermion superconductor UTe2

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Recently, UTe2 was newly discovered as a candidate material of a spin-triplet superconductor. It behaves like a spin-triplet ferromagnetic superconductor below its critical temperature, exhibiting anomalously large upper critical fields. In the case of the well-studied spin-triplet ferromagnetic superconductors UGe2, UCoGe, URhGe, the Pauli depairing effect for magnetic fields perpendicular to their magnetic easy axis are suppressed, because of the exchange splitting of the Fermi surface. However, UTe2 is paramagnetic in the normal state, so the anomalously large critical field cannot be explained in the same way as other ferromagnetic superconductors. It is necessary to explore for the origin of the large upper critical fields for the perpendicular fields. For this purpose, applying the microscopic theory to this system by taking into account multi-orbital bands with higher angular momentum j=5/2, we derive the formula of the spin susceptibility and calculate it numerically. We will compare our numerical calculation results with resent experimental results of NMR Knight shift, and will discuss the Pauli limit against the field that is perpendicular to magnetic easy axis.

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Single crystal growth and de Haas-van Alphen effect of non-centrosymmetric heavy-fermion compound UPt_5

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Actinide 5*f*-electron systems show exotic phenomena such as ferromagnetic superconductivity, triplet superconductivity, and hidden order. The formation of a heavy electronic state is closely related to these exotic phenomena. UPt_5, which crystallizes in the non-centrosymmetric $AuBe_5$ structure (space group: *F*-43m (is identical to the half Heusler structure), point group: T_d^2), has been reported to have a paramagnetic heavy electronic state.[1-3] We performed de Haas-van Alphen (dHvA) experiments using high quality UPt_5 single crystals. dHvA experiment is a useful method to investigate the electronic state including the enhanced effective mass and the topology of Fermi surfaces.

We have succeeded in growing high-quality UPt_5 single crystals using the Czochralski method. UPt_5 is an incongruent melting compound, and was grown with the starting materials of an off-stoichiometric ratio. We detected dHvA oscillations, and obtained the angular dependence of dHvA frequencies and the effective mass. To interpret the observed dHvA frequencies, we performed the LDA energy band calculations. We will discuss the topology of splitting Fermi surfaces in the non-centrosymmetric crystal structure and the peculiar anisotropic effective mass in UPt_5.

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英語版に差し替え予定

空間反転対称性が破れた系の磁気構造

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Magnetic structure of system without inversion symmetry

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空間反転対称性が破れた系においては反対称スピン軌道相互作用が現れることがよく知られている。近年の技術進歩により接合系の作製において薄膜の厚さの調整や電場をかけることによって、反対称スピン軌道相互作用の強さを精密に制御できるようになった。これらの系を記述する模型として Rashba 型のスピン軌道相互作用が働く正方格子上の Hubbard 模型が考えられる。この模型において強磁性の揺らぎが発達することが報告されており[1]、発達した強磁性の揺らぎに媒介される超伝導相の実現も期待できる。実際、スピン 3 重項超伝導が安定化することが報告されており[2]。量子計算などへの応用などの観点から言っても非常に重要である。先行研究においては乱雑位相近似(RPA)を用いた解析が行われたが[1][2]、RPA では揺らぎが強く見積もられすぎることが知られているので、揺らぎ交換近似(FLEX 近似)を用いてスピン感受率の解析を行った。本講演では主たる結果を紹介する。

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Observation of Magnetoelectric Effect on Antiferromagnetic Ce Compounds with Ce Zig-Zag Chain Structure

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Ce_3TiBi_5 is an intermetallic compound with antiferromagnetic (AFM) ordering, T_N = 5.0 K. Crystal structure is the hexagonal with space group of $P6_3/mcm$ (No.193). The Ce atoms in this system form zig-zag chains along the *c*-axis. Clear cusp at T_N was observed on the temperature dependence of the magnetic susceptibility for *c*-axis, and the magnetic moment of Ce is speculated to orient along the *c*-axis below T_N. AFM ordered state on Ce zig-zag chain is equivalent to a ferroic order of magnetic toroidal dipoles on the basis of the classification of extended multipoles. Magnetization induced by electric current is expected by theoretical studies under ferrotoroidal ordered state [1]. In previous study on UNi_4B with honey comb structure, current-induced magnetization was observed below T_N [2].

We have carried out magnetization measurements under applying electric current on Ce_3TiBi_5, and have succeeded to observe a current-induced magnetization. Figure 1(a) shows temperature dependence of magnetization of Ce_3TiBi_5 at applying several current density *i*. A curve of black diamonds represents a usual magnetization of Ce_3TiBi_5 which is observed at zero electric current. Observed magnetization shows an identical temperature dependence and is not influenced by the difference of *i* above T_N. In contrast, it shows a sudden increase (decrease) with decreasing (increasing) of *i* below T_N. Deviations from the usual magnetization were extracted and are plotted in Fig. 1(b) as M_{ME}. The magnitude of M_{ME} is proportional to applied current density. In addition, we confirmed that M_{ME} is independent of magnetic field and remains even in zero magnetic field [3]. These behaviors suggest that observed current-induced magnetization is originated from the ferrotoroidal order in Ce_3TiBi_5.

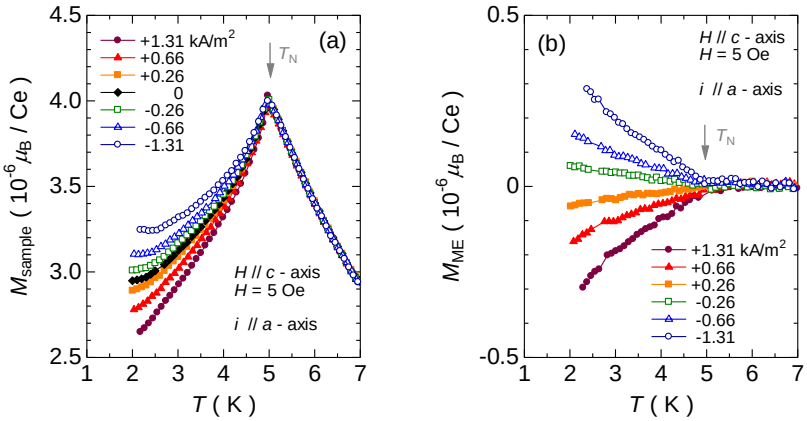

Fig. 1. Temperature dependence of (a) measured sample magnetization M_{sample} and (b) current-induced magnetization component M_{ME} on Ce_3TiBi_5 under applying several electric current.

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Novel electronic phenomena in a family of *d*-electron systems with MnP-type and TiNiSi-type structure

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We will present our systematic research for a family of *d*-electron systems which crystalize in orthorhombic MnP-type and TiNiSi-type structures in the space group *Pnma*. In these similar structures, the transition metal sites form the zigzag chain, as shown in Figs. 1 and 2, and a variety of novel electronic phenomena appears. The first feature among such phenomena in MnP-type structure is a realization of complex magnetic phase like helimagnetism, as seen in MnP, FeP, and CrAs[1]. It is thought that the crystal structure induces competition of exchange interactions among multiple magnetic sites, stabilizing helimagnetism. Second feature is an occurrence of exotic superconductivity under pressure in CrAs and MnP. They are rare examples to show superconductivity with adjoining the helimagnetic phases. Third feature is a degeneration of the Fermi surface on the Brillouin zone boundary, which is protected by the non-symmorphic symmetry. This degeneration yields novel non-magnetic phase transition in RuP and RuAs through the Fermi surface instability[2,3,4]. This crystal structure is a good stage to induce novel electronic state of both magnetic and non-magnetic origin.

To find new phase transition in the isostructural crystals, we have synthesized several compounds whose properties were not thoroughly studied before, including TiNiSi-type (CoAs, TiSe, IrSi, RhSi, TaFeGe, ZeFeGe, TaFeAs, NbCrP, NbMnP, MoFeP). We succeeded to make single crystals for some systems for the first time, and found phase transition in NbCrP and NbMnP.

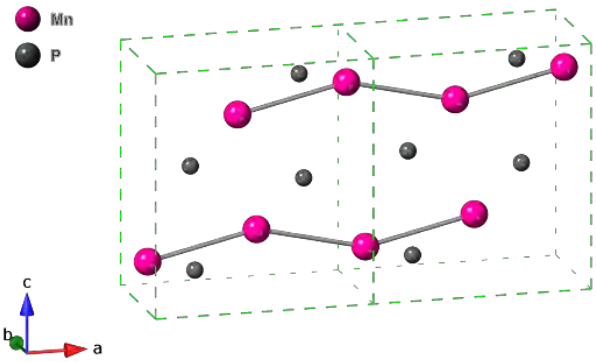

Fig.1 Crystal structure of MnP

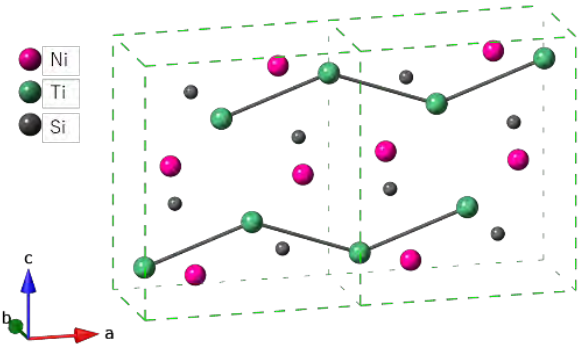

Fig.2 Crystal structure of TiNiSi

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Pressure induced quantum critical point of helimagnet Mn_3P

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Antiferromagnet Mn_3P with T_N = 30 K crystallizes in the noncentrosymmetric tetragonal structure in the I-4 space group (No. 82)[1]. The crystal structure is composed of three inequivalent Mn sites, which form tetrahedrons and zigzag chain as shown in figure.1. Since fifteen different types of Mn-Mn bonds are included within 3.0 Å, competition of the exchange interactions is expected among many Mn-Mn bonds.Our investigation revealed that this material shows a complex yet well-ordered helimagnetic structure. From electrical resistivity measurements under the pressure, the ordered state with small magnetic moments is easily suppressed by applying pressure, exhibiting a quantum critical point at ~ 1.6 GPa. Pressure-temperature phase diagram of Mn_3P is shown in the figure.2. The remarkable mass renormalization suggested by the large electronic specific heat coefficient and the large *A* coefficient of the resistivity, even in the ordered state, and an incoherent-coherent crossover in a low temperature region also characterize the unusual state in Mn_3P, which is most likely affected by the underlying frustration effect.

We will report results of NMR measurements, which are performed to study characteristic behavior in Mn_3P.

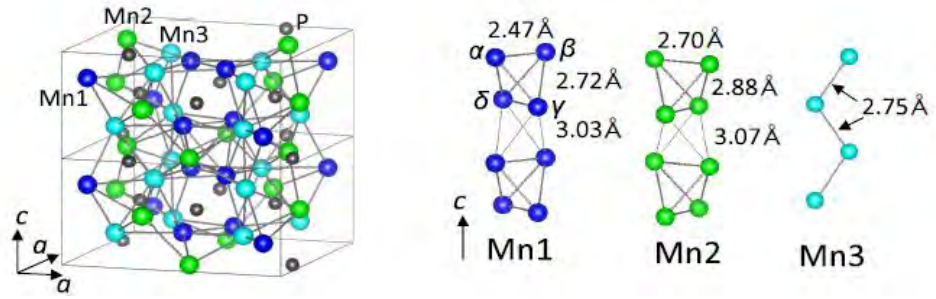

Fig. 1. Crystal structure of Mn_3P

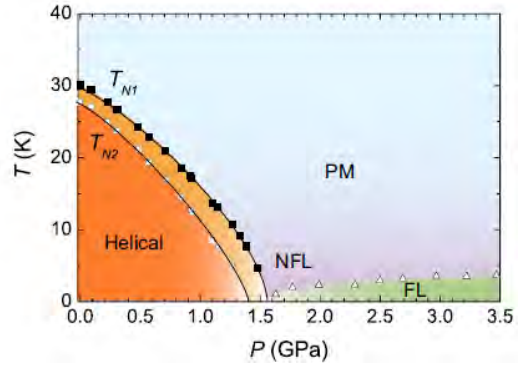

Fig. 2. Pressure-temperature phase diagram of Mn_3P

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Pairing symmetry of SrPtAs: Calculation of superfluid density and nuclear-spin-lattice relaxation rate

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The recently discovered hexagonal pnictide superconductor SrPtAs has the crystal structure with local lack of inversion symmetry [1]. There are two distinct honeycomb-shaped PtAs layers in the unit cell of SrPtAs, and the PtAs layer has checkerboard-type triangular lattice structure. Therefore, since each layer does not contain the inversion center in itself although the entire crystal is inversion symmetric, it is expected that the system has the staggered antisymmetric spin-orbit coupling. In fact, the LDA calculation of SrPtAs showed a large splitting of the bands due to antisymmetric spin-orbit coupling [2]. The LDA calculation also revealed that there are six Fermi surfaces with spin degeneracy, five of which are quasi-2D and the other 3D.

The internal spontaneous magnetization observed by the muon-spin-relaxation experiment below T_c suggests the spontaneous time-reversal symmetry (TRS) breaking in the superconducting state of SrPtAs [3]. The most probable pairing symmetry of SrPtAs suggested by the group theoretical consideration [4] and functional renormalization group (FRG) analysis [5] is the topological chiral d-wave ($d_{x2-y2}\pm id_{xy}$-wave) state with TRS breaking. On the other hand, there are still some controversies on the chiral d-wave state of SrPtAs. The nuclear spin-lattice relaxation rate measured by the nuclear quadrupole resonance showed the Hebel-Slichter peak near T_c and exponential decay in the low-temperature region [6], and the experimental results of the superfluid density exhibited the thermal-activation-type behavior at low temperatures [7]. The conventional s-wave pairing without any nodal excitation is naively expected from both the experimental results.

We calculate the superfluid density and the nuclear-spin-lattice relaxation rate based on the multiband quasiclassical formalism to address this issue, taking into account the six Fermi surfaces obtained from the LDA calculation [8]. It is shown that the recent experimental reports on the superfluid density and the nuclear-spin-lattice relaxation rate, which seemingly support the conventional s-wave pairing, are also consistent with the chiral d-wave state. We also propose that the measurement of the bulk quasiparticle density of states would be useful for the distinction between two pairing states.

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Detection of Odd-Parity Multipoles in Artificial Nanomaterials by Optical Second Harmonic Generation

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Recently, multipoles which describe electric charge and current distributions have attracted much attention from the view point of universal understanding of various physical phenomena. Particularly, magnetic toroidal dipole moment (Fig. 1), which breaks both space inversion and time reversal symmetries, has been drawing attention as an important order realizing novel magnetoelectric functionalities in multiferroics. Since the magnetic toroidal dipole moment has antiferromagnetic spin structure, however, it is difficult to directly detect it. Optical second harmonic generation (SHG) is a second-order nonlinear optical effect and sensitive to breaking of both space inversion and time reversal symmetries. Therefore, SHG can be an ideal probe to detect odd-parity multipoles such as magnetic toroidal dipole moment.

In this study, we fabricated artificial nanomaterials made by non-magnetic and magnetic materials by an electron beam lithography. Especially, triangular-shaped permalloy nanomagnets (Fig. 2) can give a valuable testing ground for the realization of mesoscale magnetic vortex states and those chiralities can be controlled by external magnetic fields due to its triangular-shape. This magnetic vortex state can be regarded as an ideal magnetic toroidal dipole moment.

For the SHG measurement, we used a Ti:Sapphire laser (central wavelength: 800 nm, pulse width: 100 fs, repetition rate: 80 MHz). Figure 3 shows magnetic field dependence of SHG intensity in increasing and decreasing runs. The difference in the SHG intensity around 0 mT comes from the opposite direction of magnetic toroidal dipole moment. Further investigations including polarization dependence of SHG intensity and symmetry analysis support our arguments. This technique is applicable to other odd-parity multipoles.

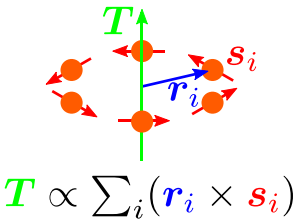

Fig. 1. Schematic image of magnetic toroidal dipole moment.

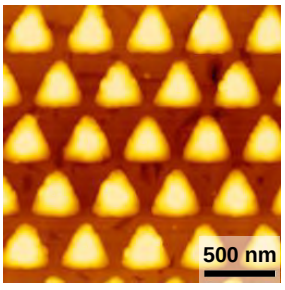

Fig. 2. AFM image of triangular-shaped nanomagnets.

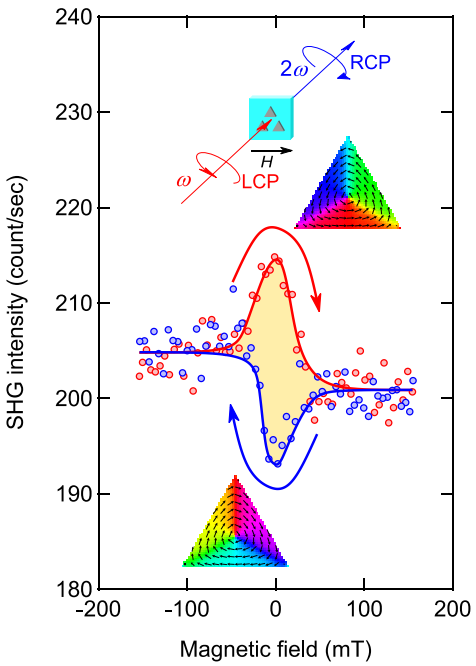

Fig. 3. Magnetic field dependence of SHG intensity in triangular-shaped nanomagnets.

B-NMR study of UNi_4B

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It has been reported that UNi_4B shows two characteristic antiferromagnetic (AFM) orderings at T_N=20.4 K and 330 mK. The crystal structure of this compound has been reported to be the CeCo4B-type hexagonal structure (P6/mmm, No.191, D^1_{6h}), where uranium atoms form triangular lattice[1,2]. The the magnetic structure is interpreted that two-third uranium moments form 60° rotated spin structure, whereas the remaining one-third uranium moments keep a paramagnetic state until T*=330 mK, below which the remaining moments order like antiferromagnetic fashion [3]. That is, the uranium magnetic moment forms a vortex-like structure with multiple propagation vector **Q** in the hexagonal (0001)-plane, so called a ferro-toroidal ordering. In such a toroidal ordering in a metallic state where the local-inversion symmetry is broken at magnetic-ion sites, it has been reported that the ME effect occurs, where a spontaneous magnetization is expected when electric current is applied for the hexagonal (0001)-plane [4]. In fact, Saito et al. succeeded to observe a current induced tiny spontaneous static magnetization of 10^{-10} μ_B/U below T_N when electric current is applied for [2-1-10] and [0001] directions [5]. However, they have pointed out the inconsistency with theoretical prediction that the current-induced magnetization is not observed for the application of the electric current parallel to [0001] direction [5]. Thus, it has been discussed that the crystal structure is not the hexagonal structure but other structure with further lower symmetry.

In order to investigate the ME effect and crystal structure, we have carried out 11B-NMR measurements for single crystalline UNi4B. ^{11}B FFT-NMR spectrum has nuclear quadrupole satellites that consists of four kinds of quadrupole split lines. Furthermore, field angle dependence of the resonance frequency in the a-plane (hexagonal [0001]-plane) obeys the two-fold rotational symmetry, evidencing that the crystal structure is orthorombic. The present NMR study demonstrates that the crystal structure of UNi_4B is not the hexagonal and it is suggested the orthorhombic crystal structure with the space group of Cmcm [6,7]. Furthermore, we have carried out B-NMR measurements with pulse current in the AFM state. When we applied field parallel to orthorhombic a-axis and current perpendicular to a-axis, NMR spectrum does not change within the experimental error. Details will be discussed in the poster session

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Study of angle dependence of thermal conductivity in the heavy fermion superconductor UTe_2

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U-based heavy-fermion superconductors have attracted much interest because of exotic superconducting (SC) properties with unusual normal states. Among these superconductors, ferromagnetic superconductors (UGe_2 [1], URhGe [2], and UCoGe [3]) are in the spotlight because of the following reasons. First, superconductivity coexists microscopically with ferromagnetism. (2) The ferromagnetic superconductors show an extremely large and anisotropic upper critical field that exceeds the Pauli limit and the absence of decrease of the Knight shift below SC transition temperature T_{SC} [4]; this implies the realization of spin-triplet pairing state. (3) Even more remarkably, the field-reinforced and re-entrant superconductivity is observed when magnetic field is applied along the hard magnetization axis [4]. Thus, these curious SC properties have been reported so far; however, the SC order parameter has not been elucidated yet because it is blurred by magnetism.

The most latest discovered U-based superconductor UTe_2 (T_{SC} ~ 1.6 K) [5], which has the characteristics similar to U-based ferromagnetic superconductor as described below, is an essential material for elucidating the mechanism of ferromagnetic superconductivity because it is a paramagnetic material located in nearly ferromagnetism, namely that superconductivity does not coexist with ferromagnetism. For example, the upper critical field, which exceeds the Pauli limit, has the strong anisotropy [5,6]. Furthermore, field-induced re-entrant superconductivity has been observed in the magnetic field along the *b*-axis (hard magnetization axis) [7,8]. One of the most important things to elucidate the SC mechanism of UTe_2 is to investigate the quasiparticle behaviours in the SC state and SC gap structure. Thermal conductivity is sensitive to delocalized quasiparticles inside SC state. So here, we performed thermal conductivity measurements for single-crystalline UTe_2 in magnetic field along the *a*,*b* and *c*-axes. In our presentation, we show the magnetic field dependence of thermal conductivity in each crystal axis direction in detail, and discuss the SC state of UTe_2 including the SC gap structure.

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Uniaxial strain effect on the $BiCh_2$-based superconductor $LaO_{0.5}F_{0.5}BiSSe$

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$BiCh_2$-based superconductors have a layered structure which is composed of $BiCh_2$-type conducting layer and blocking layer. Because of the variety of family compounds with various layered structures, the $BiCh_2$-based superconducting system is an ideal platform for the systematic investigation of the exotic superconductors. The mechanism of superconductivity was initially considered to be a phonon-mediated superconductivity from the specific heat measurement [1]. However, no isotope effect [2] and anisotropic gap from ARPES measurement [3] have been reported which suggest the possibility of unconventional superconductivity. Thus, the pairing mechanism of these $BiCh_2$-based superconductivity still remain unclear.

In the recent study of $LaO_{0.5}F_{0.5}BiSSe$ single crystal, it is reported that magnetoresistance below the onset of superconducting transition breaks the tetragonal symmetry and shows in-plane two-fold symmetric behavior [4]. This result suggests the rotational symmetry breaking in upper critical field and the possibility of nematic superconductivity. Nematic superconductivity is a novel class of superconductivity whose gap function breaks the rotational symmetry of the underlying lattice. Until now, the nematic superconductivity has been widely discussed, but many problems are still unclear. Among the most important questions is the effect of nematic superconductivity on the normal state, namely whether the superconductivity induces nematic fluctuations above the superconducting transition temperature. To investigate the anisotropic fluctuations of electronic state, elastoresistance measurement is known as a powerful probe and it is characterized by the change of resistance induced by uniaxial strain to the sample [5]. Here, we performed elastoresistance measurement in $LaO_{0.5}F_{0.5}BiSSe$ single crystal based on this technic to investigate the anisotropic fluctuation and possibility of nematic superconductivity. In the simple experimental setup for elastoresistance measurement, sample is directly attached to the surface of the piezo stack [6]. However easy this method is to control in-situ strain, it is difficult to assess the built-in strain that arise from the difference of thermal expansion between piezo stacks and ordinary materials. In general, the piezo stacks expand while ordinary material shrinks. To improve this problem, we developed the uniaxial strain apparatus that compensates the thermal expansion of piezo stack and is originally designed by C. W. Hicks *et al* [7]. Furthermore, we introduced the metal platform techniques that enable us to apply the strain to the small samples and evaluate the change in resistivity of both longitudinal and transverse direction with respect to the applied strain. In our presentation, we show the temperature dependence of elastoresistance and discuss the possibility of nematic superconductivity.

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Atomic-site analysis of new superconductor candidate $Sr_{2-x}La_xIrO_4$ by photoelectron holography using display-type Retarding Field Analyzer RFA

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The d-electron material of iridium oxide Sr_2IrO_4 has a layered perovskite type (K_2NiF_4 type) structure. This is a crystal structure similar to La_2CuO_4, one of the parent materials of copper oxide high-temperature superconductors. Due to multiple similarities, such as electronic- and atomic-structure and magnetic structure, carrier-doped Sr_2IrO_4 is expected to become a high-temperature superconductor [1, 2].

We succeeded a synthesis of iridium oxide $Sr_{1.88}La_{0.12}IrO_4$ single crystal with the world-highest electron doping (Fig. 1). In the case of such a high concentration doping materials, the position of doped atom cannot be expected simply: some La atoms might not enter the substitutional site of Sr. In the case of high concentration doped As in Si, three types of doping positions are found in XPS spectrum, and the doping site of those three components have been elucidated by the photoelectron holography technique [3]. The atomic site of doped La in $Sr_{2-x}La_xIrO_4$ single crystal has not been studied yet. Therefore, we studied the doping site of La at SPring-8 BL25SU by photoelectron holography. We used a unique two-dimensional display-type angle-resolved photoelectron analyzer RFA (Retarding Field Analyzer) [4] to measure the angular distribution of photoelectrons two-dimensionally at a time. The energy resolution is as high as 0.3 eV, which is enough to distinguish core-level-shifted peaks. Figure 2 and 3 show Sr 3d and La 4d photoelectron holograms (photoelectron diffraction patterns), respectively. Since La 4d hologram matches well with the observed and simulated holograms for Sr site shown in Fig. 2, successful analysis of La site is expected.

Fig. 1. Single crystals of $Sr_{1.88}La_{0.12}IrO_4$.

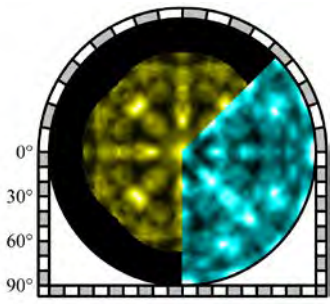

Fig. 2. Sr 3d photoelectron hologram of $Sr_{1.88}La_{0.12}IrO_4$. The right part is the simulation.

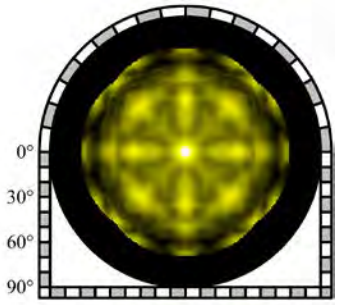

Fig. 3. La 4d photoelectron hologram of $Sr_{1.88}La_{0.12}IrO_4$.

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Hyperfine Interactions in PrT_2Al_{20} (T = Nb, Ta): ^{27}Al- and ^{93}Nb-NMR Study

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The nonmagnetic crystalline electric field (CEF) states that appear in the non-Kramers ions with an even number of *f* electrons have been attracting research interests such as nuclear refrigeration [1], enhanced nuclear magnetism [2], and multipole ordering [3]. Among them, cubic PrT_2X_{20} compounds (T = transition metal; X = Zn, Al) with a caged structure are the target of the recent experimental and theoretical studies because of their intriguing phenomena at low temperatures such as non-Fermi liquid (NFL) like behaviors and heavy-fermion superconductivity [4]. In these compounds, the interaction between conduction electrons and the non-Kramers Γ_3 doublet CEF ground state are believed to be important for the emergence of such novel phenomena. For example, both PrT_2Al_{20} (T = Nb, Ta) shows non-Fermi liquid like behaviors such as an upward curvature of the temperature dependence of resistivity below 5 K, suggesting an emergence of quadrupole Kondo effect [5].

To study phenomena related to higher-order multipoles, single crystal nuclear magnetic resonance (NMR) measurements are important because the local fields acting on nuclei give microscopic information of multipoles and *c–f* hybridizations at the ligand position. In the present study, we performed the NMR measurements on single crystalline PrT_2Al_{20} (T = Nb, Ta) and measured temperature dependence of the Knight shift for $H_0 \parallel \langle 111 \rangle$ at the Al 96*g*, Al 48*f*, and Nb sites shown in Fig. 1(a). Figure 1(b) shows the temperature dependence of the Knight shift at Al 96*g* site for both compounds. By combining with the susceptibility data, we determined the hyperfine coupling tensor at each ligand sites. In the presentation, we analyze the coupling tensor based on the spin-dipole model and discuss about the conduction bands and *c–f* hybridizations in PrT_2X_{20} compounds.

(a) (b)

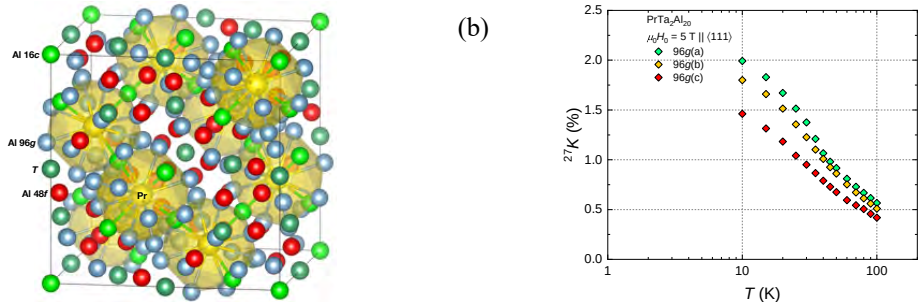

Fig. 1. (a) Crystal structure of PrT_2Al_{20}. (b) Temperature dependence of the Knight shift at the Al 96*g* site in $PrTa_2Al_{20}$.

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NMR Study on ferromagnetic superconductor UCoGe under high fields

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UCoGe is one of a few compounds in which both ferromagnetism and superconductivity originate from its U-5*f* itinerant electrons. UCoGe has strong Ising anisotropy with the *c*-axis being an easy axis and ferromagnetic (FM) fluctuations along the *c*-axis are considered to play an important role for its superconductivity, i.e. the superconductivity is induced by the ferromagnetic fluctuations[1]. In UCoGe, the superconductivity is suppressed in magnetic fields along the *c*-axis ($H \parallel c$) because $H \parallel c$ stabilizes its FM moment but suppresses its FM fluctuations. On the other hand, the superconducting (SC) upper critical field (H_{C2}) is extremely large, far beyond Pauli limiting fields in $H \parallel a$ or *b*. In particular, H_{c2} shows curious behavior when *H* is applied accurately parallel to the *b*-axis; superconductivity is enhanced by magnetic field [2]. Another interesting feature of UCoGe is 1st order FM transition reported by the nuclear quadrupole resonance (NQR) measurement in zero field, although FM transition is 2nd order in most ferromagnets [3].

In order to understand intriguing superconducting and magnetic properties of UCoGe in $H \parallel b$, we performed ^{59}Co-NMR measurement up to 23 T, beyond the FM critical field where FM order is suppressed. Because the nuclear spin-lattice relaxation rate $1/T_1$ of UCoGe is strongly suppressed by the *c*-axis magnetic field component, we have to control the angle between *H* and the crystalline axis of the sample with an accuracy of 0.05 degrees (Figure 1) and measured $1/T_1$ and the nuclear spin-spin relaxation rate $1/T_2$. We investigated the field dependence of magnetic fluctuations in $H \parallel a$ and $H \parallel b$ from measurement of $1/T_1$ and $1/T_2$. We also measured temperature dependence of NMR spectra and found clear 1st order behaviors at T_{Curie} even in magnetic field precisely parallel to the *a*- or *b*-axes.

In our presentation, we will show the relationship between superconductivity and FM critical fluctuations and compare with the case of URhGe [4], which is a sister compound of UCoGe and shows the enhancement of superconductivity in high field along to the *b*-axis. Moreover, we will also discuss the magnetic properties of UCoGe from NMR spectra.

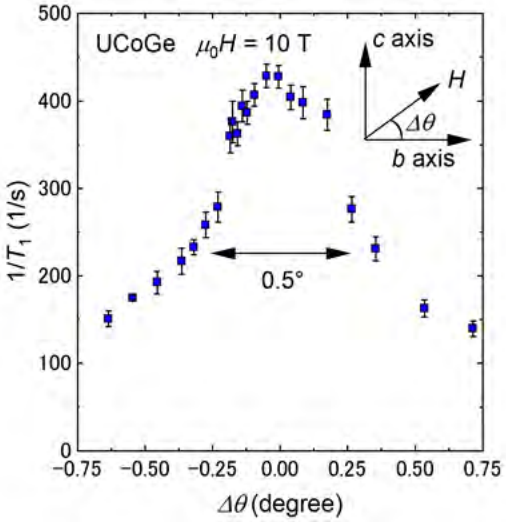

Figure 1: Angle dependence of $1/T_1$ in the single-crystal UCoGe. $\Delta\theta$ is angle between the *b*-axis and the field in the *bc*-plane.

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