

Discovery of spin-vortex-crystal magnetic order in Ni- and Co-doped $\text{CaKFe}_4\text{As}_4$

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In this presentation I will review or recent work [1-5] on the growth and characterization of pure and Co- or Ni-substituted $\text{CaKFe}_4\text{As}_4$. We find that, with pressure, a series of two half collapsed tetragonal phase transitions take place[3,4], the first across the Ca layer, destroying the superconducting state. We find with Co- or Ni-substitution that a new magnetic state is stabilized, a spin-vortex-crystal (SVC) state [5]. The SVC state that does not have any structural phase transition associated with it. Details of both crystal growth [1,2] as well as determination and implication of the SVC state [5] will be given.

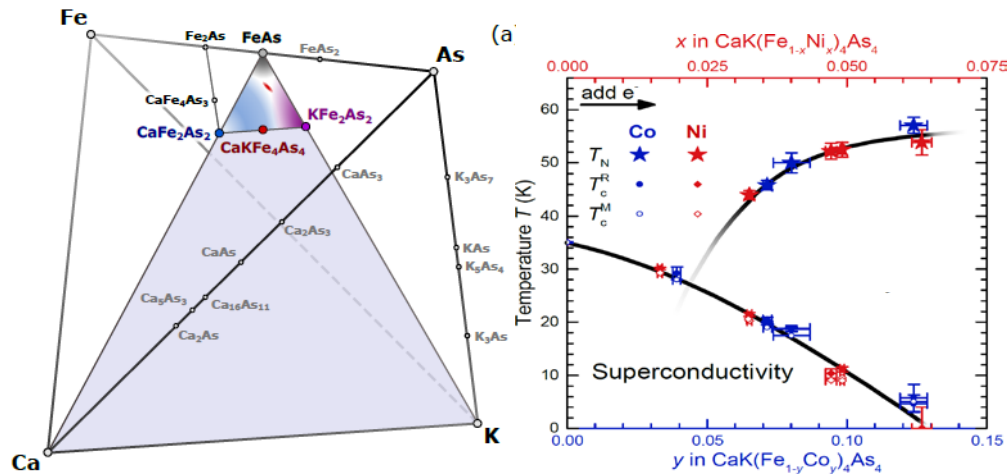


Fig. 1. (left figure) Quaternary phase diagram for growth of $\text{CaKFe}_4\text{As}_4$ [2]; (right figure) Transition temperature versus substitution level for Co- and Ni-substituted $\text{CaKFe}_4\text{As}_4$. [5]

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New Strategies for designing bismuth chalcogenide layered superconductors

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Since the discovery of the BiS₂-based superconductor in 2012 [1,2], many kinds of BiS₂-based superconductors have been discovered and the record of transition temperature (T_c) exceeded 10 K [3]. The typical structure of the BiS₂-based superconductor is represented as an alternate stacks of a conducting BiS₂ layer and an insulating (blocking) layers such as a REO layer (RE: La, Ce, Pr, Nd, Sm, etc.). Basically, the parent phase is an insulator with a band gap, and electron-doped phase shows superconductivity. However, in a typical system RE(O,F)BiS₂, both carrier doping and the optimization of the local crystal structure were found to be essential for the emergence of bulk superconductivity [4]. To develop superconducting materials with BiS₂-type layers, new material design concept is desired.

Recently, we successfully analyzed the crystal structure of La₂O₂Bi₂Pb₂S₆ [5,6], which is new type of layered bismuth chalcogenide: Bi and Pb are selectively occupy the BiS₂-like layer and the PbS-like layer, respectively. Namely, the structure can be regarded as an alternate stacks of the LaOBiS₂-type layer and the rock-salt-type PbS layer. In addition, on the basis of this stacking concept, we newly synthesized La₂O₂Bi₃AgS₆, in which the Ag_{0.5}Bi_{0.5}S is inserted into the van-der-Waals gap of LaOBiS₂ [7]. We will show the synthesis, crystal structure, and physical properties of new La₂O₂M₄S₆ (M = Bi, Pb, Ag, Cd) and discuss the possibility of superconductivity in this new layered oxychalcogenide system.

In addition, we recently synthesized new RE(O,F)BiS₂ with a high-entropy-alloy-type (HEA-type) blocking layer [8]. The REO layer is designed by the concept of HEA [9]. With HEA-type blocking layer, superconducting properties seem to be improved as compared to normal system with one or two RE elements only. We will briefly show the material design concept and latest results.

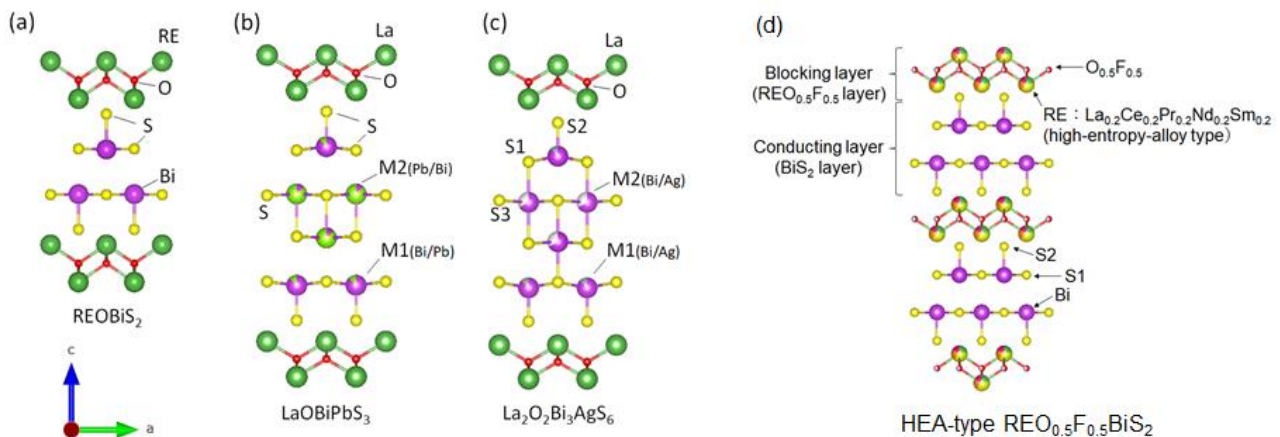


Fig. 1. Crystal structure images of (a) REOBiS₂, (b) LaOBiPbS₃, (c) La₂O₂Bi₃AgS₆, (d) HEA-type REO_{0.5}F_{0.5}BiS₂.

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Tunable superconductivity and magnetism in topological semimetal candidate RPdBi

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Topological insulators provide a new type of classification for quantum phases of matter, characterized by topological invariants [1]. Unlike traditional phase transitions described by the concepts of symmetry breaking, topological order emerges even in the absence of symmetry breaking. Combined with symmetry-breaking ordered states, topological order can give rise to unusual collective modes, such as Majorana fermions with superconductivity and axions with magnetic order [2,3].

The large family of ternary half-Heusler compounds is a prime candidate for the combination of topological and symmetry-breaking phases to realize new collective excitations [4]. These materials are located at the border between topologically trivial and nontrivial electronic band structures, which can be tuned via atomic number and/or spin-orbit coupling strength. Besides, in the rare earth-based half-Heusler series, superconductivity and magnetism can be stabilized at low temperatures, indicative of a promising platform to investigate the interplay between symmetry breaking and topological order.

We here report a systematic study on superconductivity and magnetism in a new family of topological semimetals, the ternary half-Heusler compound RPdBi (R: rare earth). We find that controlled by substitutions of rare earth elements, fine-tuning the de Gennes factor scales the Neel order while simultaneously suppressing the superconductivity as shown in Fig.1 [5]. The combination of magnetism and superconductivity, together with tunable band inversion strength associated with topological order in this system, provides a unique route to realize novel quantum states of matter.

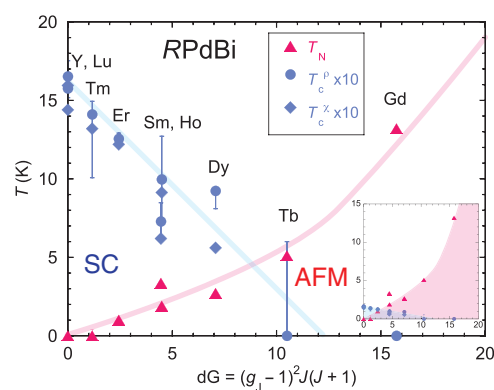


Fig. 1. Superconducting transition and Neel temperatures for RPdBi as a function de Gennes factor $dG = (g_J - 1)^2 J(J + 1)$ [5].

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Superconductivity and polymorphism in hexagonal BaPtAs with an ordered honeycomb network

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Alkaline-earth platinum pnictides exhibit a variety of hexagonal structures that are characterized by honeycomb networks, such as $\text{CaPt}_x\text{P}_{2-x}$, SrPtAs , and BaPtSb with an AlB_2 -($P6/mmm$, D_{6h}^1 , No. 191), a KZnAs -($P6_3/mmc$, D_{6h}^4 , No. 194), and a SrPtSb -type ($P-6m2$, D_{3h}^1 , No. 187) structures, respectively. SrPtAs exhibits superconductivity at 2.4 K, as we reported [1]. Superconductors with honeycomb networks have attracted interest since the theoretical predictions of exotic superconductivity in SrPtAs , such as a singlet-triplet mixed state [2], a chiral d -wave state [3], and an f -wave state [4]. In order to explore the exotic superconducting states, we have developed novel compounds with honeycomb networks.

BaPtAs has been known to crystallize in the cubic LaIrSi -type structure ($P2_13$, T^4 , No. 198). We discovered novel hexagonal structures of BaPtAs with ordered PtAs honeycomb networks, namely, SrPtSb - ($P-6m2$, D_{3h}^1 , No. 187) and YPtAs -type ($P6_3/mmc$, D_{6h}^4 , No. 194) structures [5]. Both phases exhibited superconductivity at 2.8 and 2.1–3.0 K, respectively [5]. Moreover, we also discovered superconductivity at 1.64 K in BaPtSb with the SrPtSb -type structure [6]. Inversion symmetry is broken in the SrPtSb -type, whereas it is preserved in the YPtAs -type. Our discovery provides opportunities not only for the experimental examination of the predicted superconductivity but also for further studies on exotic states that result from the strong spin-orbit interaction of Pt under broken inversion symmetry.

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Superconducting properties and anisotropic superconducting gap of CeIr₃ single crystal

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The exploration of superconducting properties in Ce-based systems has been one of the most intriguing subjects in the research field of unconventional superconductivity[1]. In this study, we aim to elucidate superconducting properties of CeIr₃, which crystallizes in the PuNi₃-type rhombohedral structure with the space group R-3m (#166, D_{3d}^5). There are two crystallographically non-equivalent Ce sites (Ce1 and Ce2) and three Ir sites, as shown in Fig. 1. The superconductivity with a transition temperature of 3.34 K in CeIr₃ was first reported by Geballe *et al.* in the 1960s[2]. Previous studies were performed at zero field using polycrystalline samples, and the thermodynamic bulk properties have not been reported so far.

Single crystals of CeIr₃ were grown by the Czochralski method in a tetra arc furnace under an argon atmosphere. CeIr₃ has been reported to be an incongruent melting compounds. We tried the Czochralski method using starting materials with an off-stoichiometric ratio and succeeded in growing single crystals. We obtained single crystals with dimensions of approximately $1.5 \times 1.0 \times 1.0 \text{ mm}^3$, as shown in the inset of Fig. 2. The superconducting properties of CeIr₃ single crystal are investigated by electrical resistivity, specific heat, and magnetization. Figure 2 shows the temperature dependence of specific heat in the form of C/T and exhibits a clear jump corresponding to the superconducting transition at 3 K, indicating bulk superconductivity in CeIr₃. We will discuss the superconducting properties and the superconducting gap structure of CeIr₃ single crystal in the presentation.

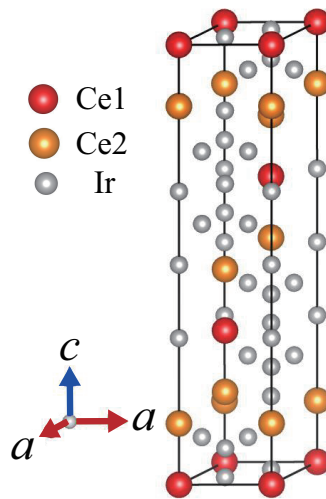


Fig. 1. Rhombohedral crystal structure of CeIr₃.

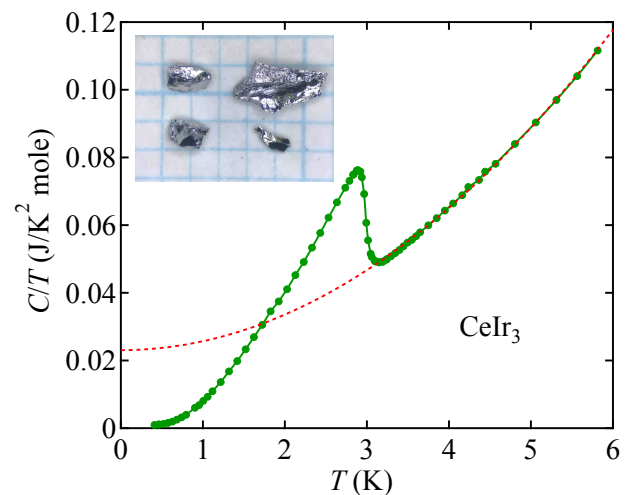


Fig. 2. Temperature dependence of the electronic specific heat of CeIr₃ single crystal. The inset shows pictures of CeIr₃ single crystal.

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Novel topological phases in strongly correlated d- and f-electron materials

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The prediction and discovery of new topological states of matter and their effects on the bulk properties of materials has attracted widespread attention [1,2], as they are promising candidates for future technological applications such as quantum computing, memory storage, and sensors. While much of the research has focused on the exploration of topological states in materials without strong electronic correlations (e.g. Bi₂Se₃), these correlations open up new routes to generating novel topological states. For example, the competing interactions among the magnetic moments of d- or f-electrons leads to magnetic frustration and often gives rise to non-collinear or non-coplanar spin structures. Mobile conduction electrons feel the effects of a large (fictitious) magnetic field when they move in the topological spin texture of these non-collinear and non-coplanar magnets, which gives rise to a large anomalous Hall effect in compounds such as Mn₃Sn [5]. Furthermore, the conducting surface state in the Kondo insulator SmB₆, produced from strong hybridization of the Sm 4f electron and conduction electron states, may be a new example of a topological state in a correlated electron material [3,4]. Actinide compounds, with their larger overlap of the f-electron orbitals with neighboring ligand orbitals, often have larger characteristic energy scales and provide fertile ground for searching for new and interesting topological materials. In this talk, I discuss our recent work on the topological properties of the PuB₄ insulator and the non-collinear antiferromagnet Mn₃Sn.

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Quasilinear quantum magnetoresistance in pressure-induced nonsymmorphic superconductor chromium arsenide

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Linear magnetoresistance is a subject of high interest. In recent years, linear magnetoresistance has been observed in many Dirac materials, in which the electron-electron correlation is relatively weak. The strongly correlated helimagnet CrAs undergoes a quantum phase transition to a nonmagnetic superconductor under pressure [1, 2]. We have measured the transverse magnetoresistance of CrAs under pressure [3]. In the pressure range close to where the superconducting transition temperature is maximised, our low-temperature magnetoresistance exhibits a striking non-saturating, quasilinear magnetic field dependence up to 14 T. Our bandstructure calculations reveal a subtle band crossing near the Y-point of the Brillouin zone, which is protected by the nonsymmorphic crystal symmetry. In this presentation, I will show that the quasilinear magnetoresistance arises from an intricate interplay between the nontrivial band crossing and strong magnetic fluctuations.

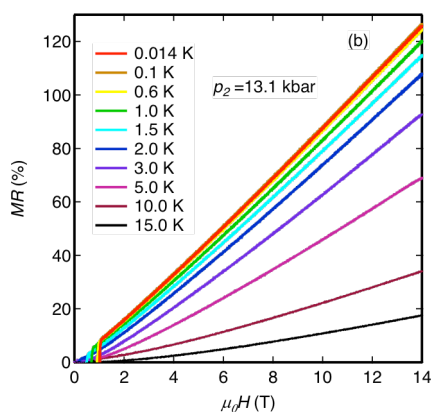


Fig. 1. Magnetoresistance of CrAs at 13.1 kbar.

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Magnetocaloric effect and its implementation in critical behavior study of the ferromagnet Nd₂Pt₂In

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Magnetocaloric effect (MCE) and critical behavior study in the ferromagnet Nd₂Pt₂In has been investigated based on the isothermal magnetization measurements. The ferromagnetic (FM) transition has been observed at $T_C = 16$ K from the resistivity, magnetic susceptibility and heat capacity measurements. Large MCE around T_C are found to be 6.24 J/(kg.K) for a field change of 7 T with a relative cooling power (RCP) value of 116 J/kg. The Arrott – plots and universal curves of the rescaled ΔS_M confirm that the magnetic phase transition in Nd₂Pt₂In compound is of the second – order in nature. The field dependence of the magnetic entropy change $(-\Delta S_M|_{T_C} \sim (\mu_0 H)^n)$, n being the critical exponent) has been used and implemented to obtain the critical exponent value of $n = 0.65$. The critical behavior study indicates that Nd₂Pt₂In has two different behaviours below and above T_C . Below T_C , it closes to the 3D – Heisenberg model and above T_C it follows the 3D – Ising model.

Quantum criticality in antiferromagnet Mn_3P under pressure

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Recent discovery of superconductivity in MnP at a magnetic phase boundary has motivated the exploration of quantum phase transition in Mn-based systems.[1] However, Mn-based compounds generally tend to have strong magnetic interaction and the ground states of most of Mn-based compounds are magnetically-ordered state with a large magnetic moment. In this presentation, as a rare example, we will show that a magnetic quantum phase transition occurs in Mn_3P under a relatively lower pressure.

Mn_3P crystalizes in a tetragonal structure as shown in Fig.1 and the space group is I-4. The Mn ions have three inequivalent sites, and the space inversion symmetry is lacking. The antiferromagnetic transition occurs at $T_N = 30$ K, [2] but a magnetic structure has not been clarified yet. We synthesized single crystals of Mn_3P using a flux method. Figure2 shows temperature dependence of resistivity at ambient pressure. The resistivity shows a clear hump below T_N and possesses the relatively high residual resistivity, indicative of a gap formation by the antiferromagnetic transition. This behavior resembles that of $\alpha\text{-Mn}$. [3] By applying pressure, the transition is gradually suppressed, accompanied by the decrease in the residual resistivity, and the ordered state disappears at 1.5 GPa. In the vicinity of quantum critical point, the behavior in resistivity is similar to those of heavy fermion systems, and the A coefficient in resistivity shows high value among d electron systems.

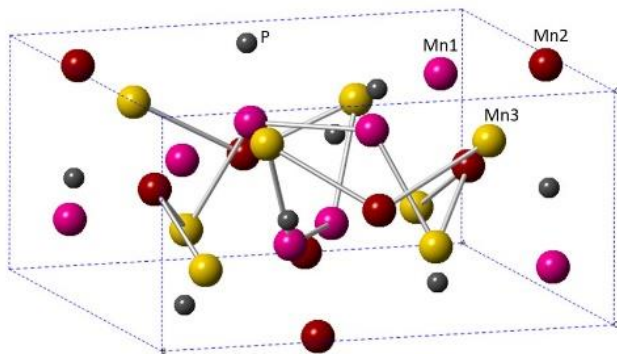


Fig. 1: Crystal structure of Mn_3P

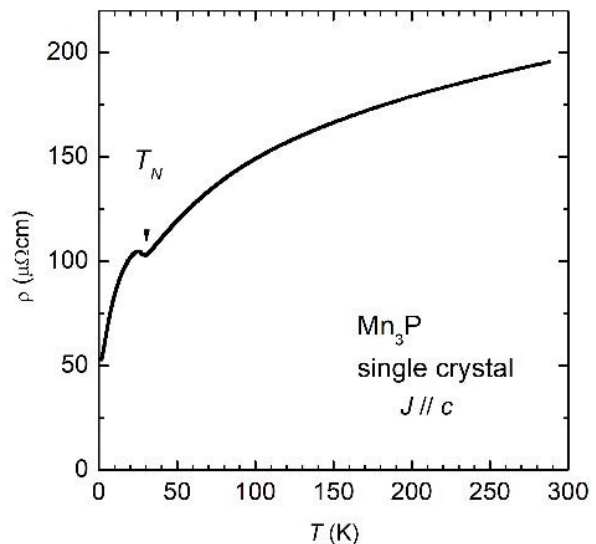


Fig. 2: T dependence of resistivity of Mn_3P at ambient pressure

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New puzzling results on the valence, the magnetism, and the superconductivity in Kondo lattice systems

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Taking profit of the huge improvement in resolution provided by modern resonant X-ray emission spectroscopy techniques, we studied the temperatures dependence of the 4f occupancy in a series of Yb Kondo lattices covering a wide range of Kondo temperatures T_K [1]. The Kondo model predicts that both the valence at low temperatures and its temperature dependence should scale with the characteristic energy T_K . Indeed we observed a nice scaling between the valence at low temperature and T_K obtained from thermodynamic measurements, in agreement with simple theoretical models. However, the temperature scale T_v at which the valence increases with temperature is almost the same in all investigated materials despite the Kondo temperatures differ by almost four orders of magnitude. This observation is in remarkable contradiction to both naive expectation and precise theoretical predictions of the Kondo model, asking for further theoretical work in order to explain our findings. Further on, our very precise data on the alloy system $\text{Yb}(\text{Rh}_{1-x}\text{T}_x)_2\text{Si}_2$ with $T = \text{Co, Ir}$ show the same temperature dependence across the whole series, without any evidence for a critical behaviour. This provides a very strong indication against a proposed quantum critical valence transition in YbRh_2Si_2 .

I shall further on present a new system close to a possibly quadrupolar quantum critical point, and, if time allows, discuss proposed superconducting order parameter for CeCu_2Si_2

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Coexistence of Superconductivity and Antiferromagnetism in Heavy-Fermion Systems with Multiple Kondo Sites

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Superconductivity (SC) emerging in some heavy-fermion compounds at the verge of magnetic instability continues to be one of the most intriguing yet still unsolved central problems of modern condensed matter physics. In our on-going research we address the issue of the coexistence of SC and long-range antiferromagnetic (AFM) ordering in crystalline materials bearing multiple inequivalent lattices of localized magnetic moments [1]. The specific systems of our interest are $\text{Ce}_3\text{PdIn}_{11}$ and $\text{Ce}_3\text{PtIn}_{11}$, which possess two Kondo sublattices in their crystal structure [2]. In both indides, heavy-fermion SC sets in within an AFM state, and their phase diagrams indicate the presence of quantum critical points induced by hydrostatic pressure or/and external magnetic field [3,4]. In my talk, I will present our recent experimental results, and discuss possible interdependence of the two cooperative phenomena occurring despite their formal separation in the reciprocal space.

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Unusual Heavy Electron State in Sm- and Yb-based 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. Cage compounds with the general chemical formula RTr_2X_{20} (*R*: rare earth, *Tr*: transition metal, *X*: Al, Zn, and Cd) crystallize in a cubic structure with the space group $Fd\bar{3}m$. Remarkable behaviours found in this series include an HF state with extremely large Sommerfeld coefficient of $\gamma = 8 \text{ J}/(\text{mol} \cdot \text{K}^2)$ in $\text{YbCo}_2\text{Zn}_{20}$ and a nonmagnetic Kondo effect and quadrupole fluctuation mediated superconductivity in $\text{PrTr}_2\text{Al}_{20}$ (*Tr*: Ti and V).

In the $\text{SmTr}_2\text{Al}_{20}$ family (*Tr*: Ti, V, Cr, and Ta), unusually field-insensitive phase transition [$T_x=6.5 \text{ K(Ti)}$, 2.9 K(V) , 1.8 K(Cr) , and 2.0 K(Ta)] and HF state have been discovered. The CEF ground state of these Sm compounds is expected to be a Γ_8 quartet of the $J = 5/2$ multiplet. Since the size of the ordered moment is extremely suppressed compared with that expected for a Γ_8 ground state, the multipolar degrees of freedom included in the Γ_8 state are expected to play some role in this field-insensitive ordered state. The magnetic susceptibility of the compound shows weak temperature dependences, which are evidently different from those for both free Sm^{2+} and Sm^{3+} ions. X-ray absorption spectroscopy (XAS) studies revealed that the Sm ions in the $\text{SmTr}_2\text{Al}_{20}$ family are in mixed valence states with an average Sm ion valence of 2.87 with no significant T dependence between 7 and 300 K[2,3]. These results suggest that valence fluctuation play an important role in the field-insensitive nature. We would like to discuss all these experimental results on SmTr_2X_{20} and recent progress of theoretical works considering valence fluctuation [4].

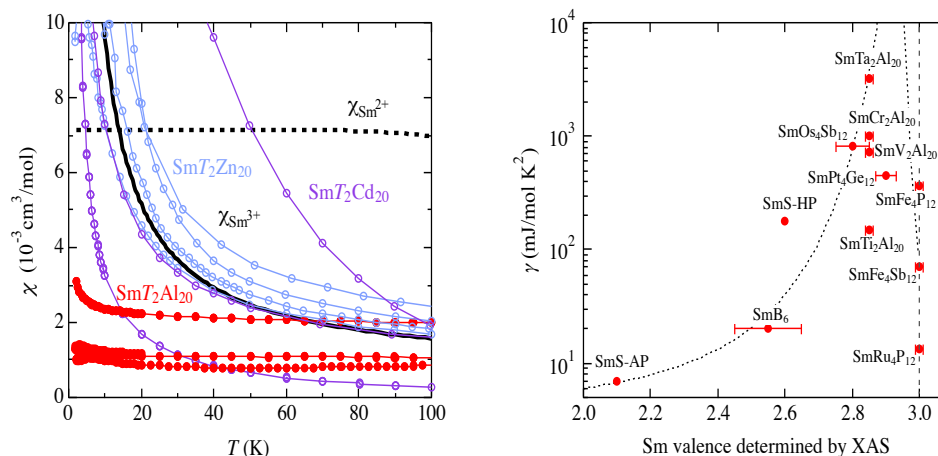


Fig. (a) Magnetic susceptibility of SmTr_2X_{20} , (b) Sm valence determined by XAS vs Sommerfeld coefficient γ value on Sm-based compounds

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Single-site non-Fermi liquid behaviors in a diluted Pr system $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$

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Non-Fermi liquid (NFL) behaviors of a U or Pr ion with f^2 configuration have received considerable attention since single-site quadrupole Kondo effect was proposed to describe the NFL behaviors of a heavy fermion compound UBe_{13} [1]. Up to now, however, the single-site quadrupole Kondo effect has not been established because of inevitable atomic disorder and/or uncertainty of crystalline electric field (CEF) levels of the f^2 electrons. Recently, the NFL behaviors of the specific heat C and the electrical resistivity ρ were observed in a cubic $\text{PrIr}_2\text{Zn}_{20}$ with the Γ_3 doublet ground state carrying active quadrupoles, suggesting formation of the quadrupole Kondo lattice [2, 3]. If this is the case, the single-site quadrupole Kondo effect could manifest itself when the Pr^{3+} ions are diluted with elements without $4f$ electrons. Bearing this in mind, we have measured C and ρ of a diluted Pr^{3+} system $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$.

The magnetic specific heat divided by temperature C_m/T for $x \leq 0.44$ is divergently enhanced on cooling for $T < 2$ K. A characteristic temperature of the NFL behaviors, T_0 , is defined as the temperature where the magnetic entropy S_m reaches 75% of $R\ln 2$. As displayed in the inset of the figure, the obtained T_0 for $x = 0.024$ as 0.28 K is monotonically increased to 1.37 K for $x = 0.44$. As shown in the figure, the data of C_m/T for all x normalized by the absolute values at T_0 converge well at $0.5 \leq T/T_0 \leq 3$. Moreover, the differential electrical resistivity $\Delta\rho (= \rho(T) - \rho(3\text{ K}))$ normalized by T_0 converges at the similar temperature region. The scaling of both C_m/T and $\Delta\rho$ data indicates the NFL behaviors originate from an identical mechanism. Furthermore, for $x < 0.05$, the normalized C_m/T and $\Delta\rho$ data follow $-\ln T$ and $\sqrt{T}$ at $T/T_0 \leq 1$, respectively [4]. The temperature dependences are consistent with those expected from the single-site quadrupole Kondo model [1]. Therefore, we infer that the observed NFL behaviors result from the single-site quadrupole Kondo effect. On the other hand, the normalized data of C_m/T for $x = 0.085$ and 0.44 deviate from the $-\ln T$ dependence at $T/T_0 \leq 0.5$, which suggests effect of atomic disorder and/or an inter-site interaction between Pr ions on the ground state.

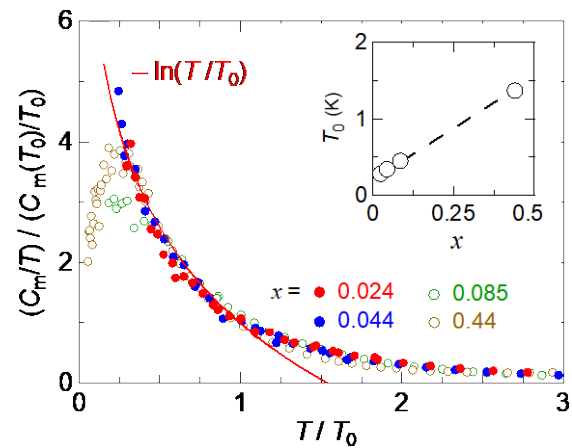


Fig. Scaling plot of the magnetic specific heat divided by temperature C_m/T in $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$ for $x \leq 0.44$. The inset shows the x -dependence of the characteristic temperature T_0 of the NFL behaviors.

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Current-induced magnetization on Ce-based antiferromagnetic metals

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Recent intensive theoretical studies revealed that various phenomena such as magnetoelectric (ME) effect can be well understood and categorized on the basis of spatially extended multipoles [1,2]. In addition, it is also predicted that ME effect can occur even in the metallic system, although ME effect have discussed mainly in insulating system so far. It is suggested in one of the above theories [3] that an antiferromagnetic (AF) order at $T_N = 20.4$ K on UNi₄B can be described as a ferroic order of toroidal moment $\mathbf{t}$, so that it may show predicted ME effect. Toroidal moment is one of the odd parity multipole and is sometimes used to describe magnetoelectric coupling. Thus, in order to test the possibility of the theoretical predictions, We have performed magnetization measurement under direct electric current on UNi₄B. We observed that magnetization is induced by constant electric current in the ordered state of UNi₄B [4]. Therefore, the validity of the theory is confirmed in part by the experiments. However, the fact that the magnetic structure of UNi₄B is not fully confirmed makes it difficult to compare the experimental results with the theory, hindering further understanding of the observed phenomenon.

We have recently started searching for another candidate metal for the theory. In this presentation, we report two collinear antiferromagnetic systems which show current-induced magnetization, CeRh₂Si₂ and CeRu₂Al₁₀, and discuss about the cause of this phenomena.

CeRh₂Si₂ crystallizes into ThCr₂Si₂-type tetragonal structure (symmetry: I4/mmm, D_{4h}¹⁷, No.139). It shows successive antiferromagnetic transitions at $T_{N1} = 36$ K and $T_{N2} = 25$ K. These orders possess the wave vector $\mathbf{q} = (1/2, 1/2, 0)$ and $\mathbf{q} = (1/2, 1/2, 1/2) + (1/2, 1/2, 0)$, respectively. Magnetic moments are oriented along the [001] direction in both phases [5]. We found that magnetization in the direction of c axis is induced by the application of direct electric current along a axis only between T_{N1} and T_{N2} .

CeRu₂Al₁₀ crystallizes into YbFe₂Al₁₀-type orthorhombic structure (symmetry: Cmcm, D_{2h}¹⁷, No.63). Ce ions align along c axis forming zigzag structure. Ce ions participate an antiferromagnetic order at $T_N = 27$ K. This order is characterized by a propagation vector $\mathbf{q} = (0, 1, 0)$ where magnetic moment align along c axis [6]. Single crystalline sample is grown by H. Tanida (Toyama Pref. Univ.). We found magnetization is induced by electric current below T_N for the settings of ($\mathbf{I} \parallel a$, $\mathbf{B} \parallel c$) and ($\mathbf{I} \parallel c$, $\mathbf{B} \parallel a$ and b).

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X-ray Crystal Structure Analysis of Toroidally Ordered System UNi₄B

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The interplay between electric and magnetic properties of electrons in matter has attracted much interest in recent condensed matter physics. In particular, the phenomena that magnetism couples with ferroelectricity, which is called the magnetoelectric (ME) effect, has been studied intensively since the discovery of multiferroic materials. The ME effect had been considered as a property that only insulating materials can exhibit because metallic materials have no electric polarization. However, a recent theoretical study proposes that the ME effect can arise also in metals in which the magnetic moments order in vortex-like arrangements, referred to as toroidal order [1]. UNi₄B is the first example of toroidal ordering metal that is confirmed experimentally to exhibit magnetization induced by electric current [2]. The experiment and theory are, however, not fully consistent with each other about the directions of applied electric current and induced magnetization. One of the problems that hinder understanding of this phenomena is the crystal structure of this compound. There are two controversial reports of the crystal structure of this compound: the hexagonal (*P6/mmm*) [3] and orthorhombic (*Cmcm*) [4] ones, both of which were investigated using laboratory X-ray sources. We performed crystal structure analysis using high-energy synchrotron X-ray, in order to determine the crystal structure of UNi₄B.

A small piece (about 20 microns in diameter) of single crystalline UNi₄B grown by Czochralski method was used as a specimen. The diffraction of synchrotron X-ray was measured by using a diffractometer with an imaging-plate type detector at a beamline PF-AR NE1A in Photon Factory. The high energy X-ray of 30 keV was utilized to reduce the large absorption effect of the heavy U atoms in the sample. The obtained diffraction patterns strongly suggest the orthorhombic unit cell, whose lattice constants are: $a = 6.922(4) \text{ \AA}$, $b = 14.773(2) \text{ \AA}$, $c = 17.04(1) \text{ \AA}$, which is the same as one of the previously reported structures [4]. The observed reflection conditions indicate possible space groups as follows: *Cmcm* (D_{2h}^{17} , no. 63), *Cmc2₁* (C_{2v}^{12} , no. 36), and *C2cm* (C_{2v}^{16} , no. 40). The direct method using SIR2011 and the least-squares refinement using SHELX give very similar structures for each of these space groups, which have distorted triangular lattices formed by U atoms, without local inversion symmetry at each U site (see Fig. 1). The differences of *R*-values of the refinements among these space groups are no more than 0.5 %. In order to choose the correct space group out of these three candidates, we need a careful check from other experimental methods that are sensitive to the symmetry of the system, such as NMR, and so on.

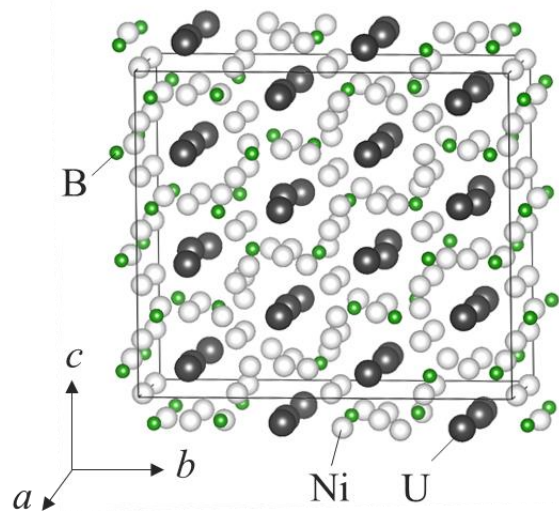


Fig. 1. Crystal structure of UNi₄B suggested by the least-squares refinement, assuming the space group of *Cmcm*.

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Carbon-based molecular materials as new electronic materials platforms

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Molecular solids whose cooperative electronic properties are based purely on π -electrons from carbon atoms offer a fertile ground in the search for exotic states of matter, including unconventional superconductivity and quantum magnetism. C_{60} -based solids with stoichiometry A_3C_{60} (A = alkali metal) are archetypal examples of molecular superconductors with the highest superconducting transition temperatures ($T_c = 38$ K) among all molecular systems [1]. We have also shown that they display the highest upper critical magnetic field ($H_{c2} > 90$ T) among all known three-dimensional superconducting solids [2]. The dominance of strong electron correlations in defining their behaviour poses significant challenges for understanding the highly robust superconducting response to both temperature and magnetic field in these highly correlated all-carbon molecular systems.

Of particular interest are also the families of molecule-based strongly correlated f -electron fullerides with stoichiometry $RE_{2.75}C_{60}$ ($RE = Sm, Yb, Eu$) in which the presence of the electronically-active C_{60} anions is combined with mixed valence rare earth ions potentially leading to properties unattainable in other systems currently available. Strong correlations dominate the electronic properties of both the rare-earth cation and the C_{60} anion sublattices and unambiguous signatures of electronically-driven valence transitions with changes in external stimuli (temperature, pressure) are found in the variation of the elastic and electronic properties of these mixed valence solids (Fig. 1).

Issues pertaining to other all-carbon π -electron systems such as the polyaromatic hydrocarbons (PAHs) will be also addressed – here claims of high-temperature superconductivity were made in materials obtained by their reaction with various metals. However, despite the flare of experimental and theoretical activity, the results have not been reproduced and the identities of all compounds in this family remained unknown, reflecting the unavailability of phase-pure samples. Recently we have been successful in devising reproducible synthetic routes of ionic salts of PAHs under mild conditions. These were shown to provide the first example of a 3D quantum spin-liquid state to 50 mK arising purely from π -electrons, while, at the same time, harboring orbitally entangled states, prerequisites of the emergence of quantum magnetism and exotic superconductivity [3,4].

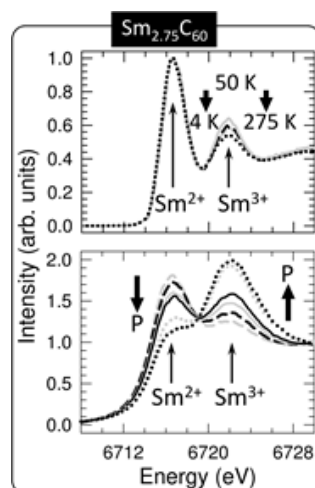


Fig. 1. Temperature- and pressure-induced valence transitions in $Sm_{2.75}C_{60}$ probed by X-ray absorption spectroscopy

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Crystal Growth of 5d Transition Metal Compounds: β -Pyrochlore Oxide CsW_2O_6 and One-Dimensional Telluride Ta_4SiTe_4

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5d transition metal compounds are of great interest in terms of the strong spin-orbit coupling effect on itinerant electron systems. I will present our recent results on exploration and crystal growth of novel 5d transition metal compounds, which may include a thermoelectric material, metal-insulator transition system, and superconducting materials.

One-dimensional telluride Ta_4SiTe_4 as a high performance thermoelectric material. Thermoelectric cooling is a promising candidate for the next-generation of refrigeration technologies in replacing vapour compression cooling using gaseous refrigerants. At present, Bi_2Te_3 -based materials are used as a practical material at around room temperature. Practical cooling at much lower temperatures enables us to refrigerate various cryogenic electronic devices and sensors. Here we report one-dimensional telluride Ta_4SiTe_4 and its substituted compounds show high thermoelectric performance at low temperature [1]. We prepared whisker crystals with typically several mm long and several μm in a diameter, shown in the left figure, and measured their thermoelectric power S and electrical resistivity ρ . S values of Ta_4SiTe_4 whisker crystals reach $-400 \mu\text{V K}^{-1}$ at 100-200 K, while maintaining low ρ of $\sim 2 \text{ m}\Omega \text{ cm}$, yielding larger power factor $P = S^2/\rho$ of $80 \mu\text{W cm}^{-1}\text{K}^{-2}$ than those in Bi_2Te_3 -based practical materials at room temperature. This very large P is probably caused by the very small spin-orbit gap opening on the strongly one-dimensional electronic bands at the Fermi energy.

Metal-insulator transition in β -pyrochlore oxide CsW_2O_6 . 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. Here we report that the β -pyrochlore oxide CsW_2O_6 , where $\text{W}^{5.5+}$ ions having $5d^{0.5}$ electron configuration form a pyrochlore structure, exhibits a unique metal-insulator transition different from other pyrochlore systems. CsW_2O_6 was found to show a metal-insulator transition accompanied by a decrease of magnetic susceptibility at around 210 K [2]. However, formation mechanism of the transition is still not fully understood, because the previous studies were employed using polycrystalline samples. We succeeded in preparing single crystals of CsW_2O_6 by a vapour transport method, as shown in the right figure. Single crystal X-ray diffraction experiments revealed that a structural change accompanied by the W_3 trimer formation, while keeping the cubic symmetry, occurs at this transition. To our knowledge, such a trimer formation has never been observed in other pyrochlore systems, indicating that a unique metal-insulator transition is realized in CsW_2O_6 .

The work has been done in collaboration with Y. Yoshikawa, T. Wada, T. Inohara, Y. Yamakawa, A. Yamakage, K. Takenaka, R. Mitoka, K. Niki, H. Amano, N. Katayama, H. Sawa (Nagoya University).

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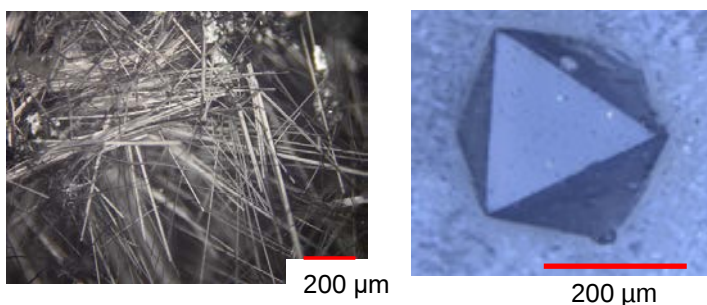


Fig. Whisker crystals of Ta_4SiTe_4 (left) and a single crystal of CsW_2O_6 (right).

Material Development Towards the Perfect Frustration on Kagome Antiferromagnet

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Geometrical frustration sometimes inhibits the development of conventional magnetic ordering, and trigger the formation of a nontrivial magnetic state. Theoretically, the realization of a spin liquid state in a quantum kagome antiferromagnet owing to the strong frustration and the quantum fluctuation has become accepted. Whereas, it is also expected an emergence of exotic magnetic states related to a higher order of spin moment, such as an octupole ordering [1], nematic phase under magnetic fields [2], *etc.* Thus, kagome antiferromagnet is one of the fascinating fields to seek interesting magnetic phenomena.

However, a lack of ideal model compounds prevents us to realize these unusual magnetic states experimentally; in actual materials, a structural distortion, an ion defect and a disorder always obscure the intrinsic magnetic properties, as pointed out in $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ (disorder), $\text{KCr}_3(\text{OH})_6(\text{SO}_4)_2$ (defect), and $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ (distortion). In order to overcome these obstacles and to observe an exotic magnetic state, especially a spin liquid state, we apply the hydrothermal synthesis to obtain new compounds in which the perfect frustration occurs on the kagome network.

We have succeeded in preparing single crystals of the $S = 1/2$ kagome lattice antiferromagnet $\text{CaCu}_3(\text{OH})_6\text{Cl}_2 \cdot 0.6\text{H}_2\text{O}$, Ca-Kapellasite [3]. This compound crystallizes in the trigonal symmetry with space group $P\bar{3}m1$, having a Kapellasite type structure. This contains a perfect kagome network of Cu^{2+} ions. The large discrepancy of the ionic radius of the non-magnetic Ca^{2+} ion and the magnetic ion prevents the ion-site mixing as observed in Zn-Kapellasite. Magnetic properties of Ca-Kapellasite were characterized by means of a magnetic susceptibility, a heat capacity, and microscopic probes on the single crystal of Ca-Kapellasite. The magnetic susceptibility revealed the compound possessed competing magnetic interactions of antiferromagnetic $J_1 = 46.8$ K, $J_3 = 12.1$ K, and ferromagnetic $J_2 = -5.3$ K. The magnetic anomaly appears only in the *ab*-plane direction at $T^* = 7.2$ K. A small peak in the heat capacity at T^* indicates the occurrence of a magnetic transition, however the quite tiny peak at T^* implies the transition is not a conventional long-range order. Remarkably, *T*-linear term 5.9 mJ/CumolK² in the heat capacity was observed in spite of the fact that the compound was an insulator. This indicates the existence of an unusual gapless excitation on the ground state of Ca-Kapellasite which is microscopically supported by the NMR and neutron scattering experiments. NMR also revealed that the spin fluctuation persisted below T^* only in the *c*-direction, while the in-plane spin moment ordered at T^* . The origin of the magnetic ordering has not been understood yet, but we expect that a novel magnetic state with spin fluctuation is realized in accordance with the strong frustration and the quantum fluctuation on kagome lattice of $\text{CaCu}_3(\text{OH})_6\text{Cl}_2 \cdot 0.6\text{H}_2\text{O}$.

In the presentation, I will talk about the magnetic properties of kagome antiferromagnet Ca-Kapellasite, details of the hydrothermal synthesis and its application for new materials searching.

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Novel Electronic Phases and Competing Interactions in the Correlated *f*-Electron Compound URu₂Si₂

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The correlated *f*-electron compound URu₂Si₂ undergoes a second-order transition at $T_0 = 17.5$ K into an ordered phase whose identity has eluded researchers for more than three decades [1]. This so-called "hidden order" (HO) phase coexists with unconventional superconductivity (SC) below $T_c \approx 1.5$ K. Application of pressure suppresses the T_c of the SC'ing phase and induces a transition from the HO phase to a large-moment antiferromagnetic (LMAFM) phase at a critical pressure $P_c \approx 1.5$ GPa [2]. Our research group found that substitution of isoelectronic Fe or Os for Ru suppresses SC and induces a transition from the HO to the LMAFM phase, similar to the behavior of URu₂Si₂ under pressure [3]. The HO-LMAFM phase transition in URu_{2-x}Fe_xSi₂ was attributed to "chemical pressure" associated with the reduction of the unit cell volume upon substitution of smaller Fe atoms for Ru atoms [3]. This allows the HO and LMAFM phases to be studied in single crystals of URu_{2-x}Fe_xSi₂ at atmospheric pressure with techniques that cannot readily be performed on URu₂Si₂ under high pressure [4] (e.g., ARPES, STM, infrared, Raman, neutron scattering, etc.). In this talk, we will review the status of ongoing research on URu_{2-x}Fe_xSi₂ single crystals in the HO and LMAFM phases, such as infrared [5] and Raman [6] spectroscopy, elastic and inelastic neutron scattering [7], electrical resistivity under pressure [8], and high field (45 T) magnetoresistance [9] measurements. Interestingly, the substitution of Os for Ru in URu₂Si₂, which also induces the HO-LMAFM transition, expands the unit cell and is inconsistent with the "chemical pressure" hypothesis, suggesting the importance of other factors (e.g., spin-orbit coupling). These investigations should be useful in developing an understanding of the underlying physics of URu₂Si₂-based materials and, perhaps, even unmasking the identity of the elusive HO phase!

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Strong Ising anisotropy of URu₂Si₂ probed by ²⁹Si NMR on the Superconducting state

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In heavy-fermion compound URu₂Si₂, unconventional superconductivity appears in the so-called ‘hidden order’ (HO) state with a superconducting (SC) transition temperature of $T_{SC} \sim 1.4$ K [1]. It is an intriguing subject to clarify the mechanism of superconductivity in the HO state and investigate the relation between HO and SC state.

For the SC gap symmetry in URu₂Si₂, the angular dependence of thermal conductivity [2, 3] and specific heat measurements [4, 5] revealed the existence of two-point nodes and a horizontal line node. For the spin part, the suppression of H_{c2} in any field direction at lower temperatures [2, 6, 7] suggests the existence of Pauli paramagnetic effects for spin-singlet pairing. However, observable change has not been detected in Knight shift (K) below T_{SC} using powder sample [8].

We have performed ²⁹Si K measurements using a high-quality single crystal of URu₂Si₂. The extremely narrow NMR spectra obtained for the single crystal allowed us to achieve much higher experimental resolution. As a result, we have obtained strong anisotropic behavior of K in the SC state, indicating that spin-singlet superconductivity is realized and that spin susceptibility in the HO state has strong Ising anisotropy [9, 10].

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Electronic structures of strongly correlated uranium compounds studied by three-dimensional ARPES

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Angle-resolved photoelectron spectroscopy (ARPES) is a powerful experimental technique capable of experimentally observing the band structure and Fermi surface of a material. It has been applied for strongly correlated *f*-electron materials, and their electronic structures have been revealed so far. On the other hand, in most of these studies, one-dimensional (1D) band structures or two-dimensional (2D) Fermi surfaces have been measured, and three-dimensional (3D) electronic structures have not been obtained. This is one of problems of these studies since most of *f*-electron materials have 3D electronic structures, and their characteristics are often not well understood.

In the present study, we have revealed the three-dimensional (3D) electronic structure of uranium compounds by 3D ARPES which is a newly-developed experimental technique. In 3D ARPES experiment, all three parameters in ARPES experiments (two detecting angles of photoelectrons and incident photon energy) are entirely scanned, and complete volume data set which cover the entire Brillouin zone is obtained. We have applied the 3D ARPES to the heavy Fermion superconductors URu₂Si₂. It was found that the U 5*f* bands are strongly hybridized with Ru 4*d* bands in the vicinity of the Fermi level. The overall 3D electronic structure was mostly explained by the band-structure calculation treating all U 5*f* electrons as being itinerant.

The work was conducted by the collaboration with Y. Takeda, T. Okane, Y. Saitoh, A. Fujimori, H. Yamagami, Y. Haga, E. Yamamoto, and Y. Ōnuki. The work was financially supported by JSPS KAKENHI Grant Numbers 16H01084 (J-Physics).

Magnetism throughout the UCoGe-URhGe-UIrGe system studied on alloy single crystals

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The ferromagnetic superconductors UCoGe and URhGe have the easy magnetization axis along the orthorhombic c -axis. Magnetization behavior in fields along b , however, plays important role in physics of these materials. URhGe at 2 K undergoes a first-order metamagnetic transition at $H_R = 12.5$ T whereas the $M(H)$ curve of UCoGe exhibits a broad S-shape around $H_m \approx 50$ T. The $\chi_b(T)$ curve of URhGe shows a sharp peak at $T_{\max} = 9.5$ K ($= T_C$) but a broad maximum around $T_{\max} = 37$ K ($\gg T_C = 3$ K) for UCoGe[1]. Similar feature – $T_{\max}$ was also detected in isostructural and isoelectronic antiferromagnet UIrGe ($T_N = 16.5$ K) along the identical axis[2, 3]. We prepared single crystals throughout $\text{UCo}_{1-x}\text{Rh}_x\text{Ge}$ and $\text{UIr}_{1-x}\text{Rh}_x\text{Ge}$ systems to establish a complex $d_{\text{U-U}}/T$ ($d_{\text{U-U}}$ is the shortest interatomic uranium distance and T is temperature) magnetic phase diagram. Our detailed study of single-crystals has revealed the evolution of the characteristic temperature ($T_{\max}$) and magnetic fields (H_R , H_m) as a function $d_{\text{U-U}}$ parameter tuned by Co/Rh and Rh/Ir alloying. This recognized a characteristic maximum in magnetic susceptibility at temperature $T_{\max}$ along the b axis as an important parameter. Three magnetically ordered regions can be distinguished within this scope; first a ferromagnetic region between UCoGe and $\text{UCo}_{0.7}\text{Rh}_{0.3}\text{Ge}$ with $T_C < T_{\max}$, second ferromagnetic region between $\text{UCo}_{0.6}\text{Rh}_{0.4}\text{Ge}$ and $\text{UIr}_{0.43}\text{Rh}_{0.57}\text{Ge}$ $T_C \approx T_{\max}$ and finally an antiferromagnetic region existing between $\text{UIr}_{0.45}\text{Rh}_{0.55}\text{Ge}$ and UIrGe with $T_N < T_{\max}$ [4] (Fig. 1).

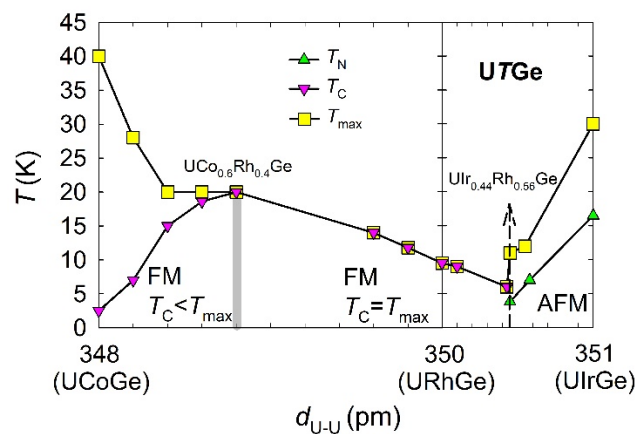


Fig. 1. $d_{\text{U-U}}/T$ phase diagram of the UCoGe-URhGe-UIrGe system.

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Single Crystal Growth and Fermi Surface Properties in Thorium Compounds

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The $5f$ -electrons in a magnetic uranium compound possess dual nature: localized and itinerant features. Namely, the $5f$ electrons contribute to the volume of the Fermi surface and also to a magnetic moment at the uranium sites [1]. A non- $5f$ thorium compound does not become a reference compound for the corresponding uranium compound because a valence of the Th atom with electron configuration $6d^2 7s^2$ are different from those of U^{3+} ($5f^3 6d^1 7s^2$) or U^{4+} ($5f^2 6d^2 7s^2$). The number of valence electrons in the thorium compound is, however, the same as that of a $4f$ -itinerant cerium compound. The topology of the Fermi surface in $ThIn_3$ [2] and $ThRu_2Si_2$ [3] is approximately the same as in $4f$ -itinerant $CeIn_3$ and $CeRu_2Si_2$, for example. The cyclotron effective masses are, however, significantly different between Th compound and Ce compound. Large cyclotron masses in Ce compounds are most likely due to the many-body quantum effects, such as the Kondo effect. To clarify the electronic states of the strongly correlated nature of f -electron systems, revealing the electronic states of Th compound plays an important role.

Recently, we grew single crystal and clarified the Fermi surface and the cyclotron effective masses of $ThSb_2$, $ThBi_2$, and $ThCu_2Si_2$ by the dHvA measurement and the energy band calculations with shifted the $5f$ -partial density of states. Figure 1 is single crystals of Th compounds. In the present study, we have succeeded in growing single crystal of $ThAl_3$ with the hexagonal structure by Al-self-flux method for the first time, as shown in Fig. 1(d). The size of an obtained bar shape single crystal in $ThAl_3$ is about $1 \times 1 \times 5 \text{ mm}^3$. We will present the results of the electrical resistivity, specific heat, and dHvA effect in the presentation.

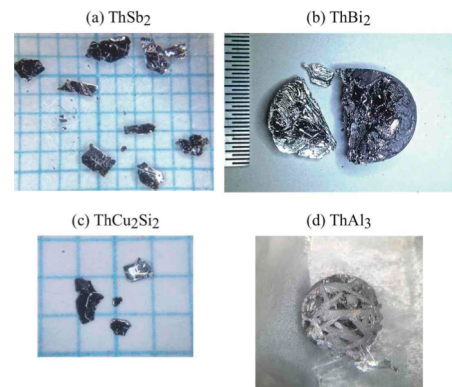


Fig. 1. Single crystals of Th compounds.

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Crystal Growth of Compounds that Contain High Vapor Pressure Element as one of the Constituents

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Crystals are the pillars of modern technology. Whether it is for basic research or for application oriented research, single crystals are very much inevitable. Although, it is very easy to prepare polycrystalline samples and perform exploratory studies, for basic research and to study the anisotropic physical properties there is always a need for single crystalline samples. In the strongly correlated electron systems, it has been well documented in the literature that high quality single crystalline samples are necessary to unravel the interesting physics in these systems. For example, several Ce-based intermetallic compounds have been tuned to quantum criticality, by means of various control parameters like pressure, magnetic field and composition [1-2]. All these experiments were performed on high quality single crystalline samples. Although there are different crystal growth techniques available, the choice of the crystal growth technique depends mainly on the melting behaviour of the compound. For example, in the case of congruently melting compounds, it is a relatively easy task to grow the single crystals directly from its melt. On the other hand, incongruently melting compounds have to be grown by dissolving them in a suitable solvent. There are certain compounds like the 122-type pnictide compounds and other Ce-based compounds that contain one of the elements which has high vapor pressure at elevated temperature, and cause difficulties in the growth of single crystals. In this talk, I will be discussing the crystal growth aspects of compounds like CeMg_3 , CaFe_2As_2 and CeTAs_2 ($T = \text{Ag}$ and Cu), where Mg and As have high vapour pressure at elevated temperature.

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Flux growth in a horizontal configuration: an analog to vapor transport growth

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Materials synthesis and crystal growth in molten fluxes have proven to be excellent routes to new materials with intriguing properties. The components of the target compound are dissolved in a flux, which is usually molten salt or metal inside of an oxide or metal crucible; crystal growth takes place under supersaturation controlled by cooling or evaporating the flux. This kind of crystal growth is normally performed in a vertical configuration. And the reaction between flux and crucible materials should be avoided as it can change the flux composition and usually leads to crystal contamination or even growth failure. In my talk, I will talk about the flux growth of novel materials in a horizontal configuration: the principles, some growth examples, and how it complements to the growth in the conventional vertical configuration.[1] I will also briefly talk about our exploratory synthesis of new superconductors by using the reaction between flux and crucible materials.[2]

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High-quality Single Crystal Growth in Heavy Fermion Compounds

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High-quality single crystals are needed to clarify the characteristic properties in heavy fermion compounds, especially the superconducting property. Cooper-pairs are broken by impurities and crystalline defects, and the existence of a residual density of states in superconductors is crucial in determining the superconducting property from the temperature dependence of nuclear spin-lattice relaxation rate $1/T_1$ below a superconducting temperature T_{sc} . Moreover, the amplitude of the de Haas-van Alphen (dHvA) oscillations, which is proportional to $\exp[-\alpha(m_c^*/H)(T + T_D)]$, is closely related to the sample quality, where $\alpha = 2\pi^2 ck_B/e\hbar$. If the cyclotron effective mass m_c^* is a rest mass of an electron m_0 , the temperature of 1 K is usual in the dHvA experiment. A much lower temperature of 0.01 K is needed for $m_c^* = 100 m_0$. Even if the low temperature is realized using a dilution refrigerator, the reduction of $\exp[-\alpha(m_c^*/H) T_D]$ is inevitable in the dHvA amplitude. Here, $T_D (= \hbar / 2\pi k_B \tau)$ is a so-called Dingle temperature, which is inversely proportional to the scattering lifetime of the carrier τ . Therefore, sample quality with a large τ value is essentially important to observe the dHvA oscillations for the heavy fermion compounds.

Followings are our typical examples in growing high-quality heavy fermion compounds[1]. These are classified into seven methods, as shown in Fig. 1, on the basis of the degree of the vapor pressure of the melt.

- 1) Czochralski method for CeCu_6 , UPt_3 , UGe_2 and CeIrSi_3
- 2) Zone-melting method for CeRu_2 and AuAl_2
- 3) Floating-zone method for CeB_6 and Sr_2RuO_4
- 4) Solid state electrotransport method for U and UPt_3
- 5) Flux method for RCu_2Si_2 , PuIn_3 , V_2Ga_5 , CoGa_3 , TiGa_3 , and ZrGa_3
- 6) Bridgman method for CePt_3Si and EuT_2X_2 (T: transition metal, X: Si, Ge)
- 7) Chemical transport method for U_3As_4 (U_3P_4), UAs_2 , UP_2 , and CoS_2

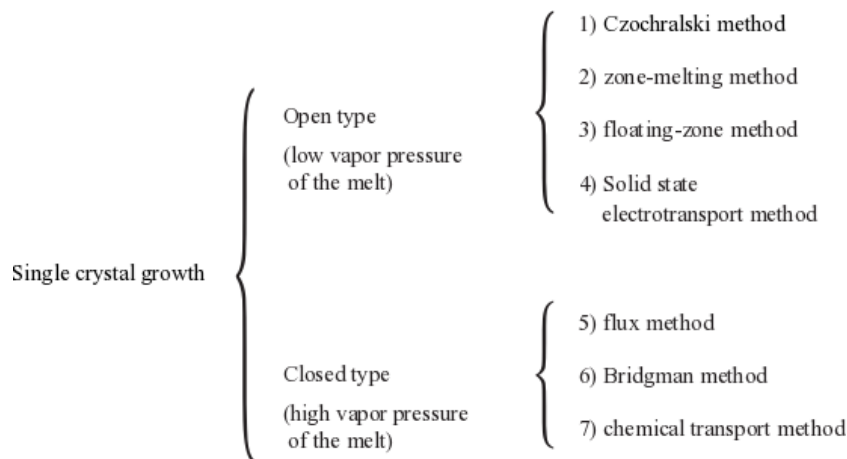


Fig. 1 Seven methods of single crystal growth.

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Crystal Growth and Physical Properties of Kondo Semiconductors

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Kondo semiconductors (KS) based on Ce and Yb have a large density of states near the Fermi energy in which a narrow gap opens with decreasing temperature below the Kondo temperature T_K [1, 2]. They attract renewed interests as candidates for correlated materials with nontrivial topology [3]. The well-known KSs are CeNiSn, CeRhSb, CeRu₄Sn₆ [4], Ce₃Bi₄Pt₃ [5], and YbB₁₂ [6], whose ground states are all nonmagnetic due to strong hybridization of the 4*f* state with conduction bands. The gap formation has been investigated by various methods using single crystalline samples. Such studies have revealed anisotropic properties in the semiconducting states as well as in the field-induced metallic states [6]. Depending on the melting point, vapor pressure, and reactivity with crucibles, we have chosen the appropriate method for the crystal growth: the rf-heating Czochralski method with an open tungsten crucible, the vertical Bridgeman method with a sealed tungsten or molybdenum crucible, and the traveling-solvent floating-zone method without any crucible in an image furnace. The homogeneity in composition was examined by the wave-length dispersive electron-probe microanalysis. Low-temperature properties of KSs are often found to be very sensitive to impurities and inhomogeneity in the single crystals [1].

In contrast to the nonmagnetic ground states in the conventional KSs, CeT₂Al₁₀ ($T = \text{Ru, Os}$) order antiferromagnetically at a rather high temperature 28 K [7,8]. Hanzawa proposed that it is related with the lack of inversion symmetry along the *b* axis at the Ce site, which allows on-site hybridization of the 4*f* state with 5*d* state [9]. Interestingly, the AFM order is preceded by the opening of a charge-density-wave (CDW) like gap in the optical conductivity along the *b* axis [10]. We have performed systematic studies of the effects of dilution and electron/hole doping on the magnetic and transport properties, electron tunneling, and spin-gap formation. It is revealed that the hybridization gap is indispensable for the unusual AFM order [11]. The fact that T_N in the isoelectronic alloy system Ce(Ru_{1-x}Os_x)₂Al₁₀ does not drop in the whole range $0 \leq x \leq 1$ indicates the importance of the half-filling condition for retaining the high T_N [12].

We have studied anisotropic dependence of the AFM transition on uniaxial pressure [12, 13]. It is found that T_N of CeT₂Al₁₀ ($T = \text{Ru, Os}$) depends linearly on the *b*-axis parameter under uniaxial pressure $P//b$ and even under hydrostatic pressure. Furthermore, irrespective of *x* in Ce(Ru_{1-x}Os_x)₂Al₁₀, application of $P//b$ markedly enhances T_N without changing T_K . This relation between T_N and T_K is in support of the model that a kind of CDW developing along the *b* axis far above T_N induces the AFM order in CeT₂Al₁₀.

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Homo-chiral crystallization and helimagnetic chirality in inorganic chiral magnetic compounds

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The chirality (left- or right-handedness) plays an important role in symmetry properties of nature at all length scales from elementary particles to cosmic science. In material science, chiral materials are often investigated in molecules or crystals with helical structures, which break mirror and inversion symmetries but combine rotational and translational symmetries. Chiral helimagnetic structure, forming only one-handed screw magnetic structure, has attracted attention because of emergence of characteristic magnetic textures such as Skyrmion lattice and chiral magnetic soliton lattice [1–2]. Therefore, it is very important to investigate relationship between crystallographic and helimagnetic chirality due to an asymmetric Dzyaloshinskii-Moriya (DM) interaction. However, there have been few experimental results due to the difficulty to synthesize the suitable materials because most of chiral inorganic compounds form racemic-twinned crystals, having both the left- and right-handed crystalline domains in a specimen.

First, our unique crystallization technique for water-soluble inorganic chiral compounds will be presented. By adapting our spontaneous crystallization technique with stirring in combination with careful examination of crystallographic chirality using X-ray, we succeeded in obtaining the centimeter-sized enantiopure single crystals of chiral helimagnetic CsCuCl₃. The large homo-chiral crystals made it possible to perform polarized neutron diffraction. The handedness of the magnetic helicity is coupled and directly controlled by the crystallographic chirality. The results could be understood in terms of DM interaction strongly coupled to the lattice [3-4]. Second, another crystal growth for non-water-soluble chiral magnetic compounds will be presented. We are developing enantiopure crystal growth techniques. We will introduce some of the techniques.

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Flux growth and characterization of new ytterbium intermetallic compounds: chiral magnetism and Kondo-lattice properties

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Flux method is a simple and useful technique to synthesize high quality crystals [1]. Because it permits the synthesis well below the melting temperature of the material, we can grow crystals which contain any high vapor-pressure element such as ytterbium (Yb). We have synthesized Yb-based intermetallic compounds using the flux method and investigated the physical properties of obtained new materials. A few years ago, we succeeded in synthesizing a new intermetallic compound of YbNi_3Al_9 using aluminum as a flux [2]. It crystallizes in a trigonal ErNi_3Al_9 -type structure with a space-group of $R\bar{3}2$ [2, 3]. This structure may have a chirality, because this space-group contains exclusively symmetry operations of the first kind, namely rotations and translations [4]. We determined the absolute structure and verified the crystal chirality using a single-crystal X-ray diffraction [5]. It is found that both left- and right-handed crystals can be obtained. Thus, YbNi_3Al_9 is suited for studying chiral magnetism in rare-earth intermetallic compound.

Recently, much attention has been given to the characteristic spin configuration in chiral magnet, such as chiral helix, skyrmion and chiral-soliton lattice [6, 7]. The chiral magnetic structure originates from an asymmetric spin interaction, so-called Dzyaloshinskii-Moriya interaction, due to spin-orbit coupling. We are interested in fundamental magnetic properties and spin-charge coupled non-trivial phenomena in YbNi_3Al_9 . To study the physical properties of YbNi_3Al_9 , we measured electrical resistivity, specific heat, magnetization, neutron scattering and hard X-ray photoemission spectroscopy. We found that YbNi_3Al_9 is a heavy fermion compound and a chiral helical magnet with the magnetic propagation vector of $q = (0, 0, 0.8)$ below the ordering temperature of 3.4 K [2, 5, 8].

Interestingly, the magnetic interaction increases by replacing Ni with Cu in YbNi_3Al_9 [5]. It is possible to substitute Cu for Ni up to 6 atomic percent by the flux method. For $\text{Yb}(\text{Ni}_{0.94}\text{Cu}_{0.06})_3\text{Al}_9$, the ordering temperature reaches 6.4 K. Using a resonant X-ray diffraction measurement, it is clarified that the crystal chirality is reflected to the helicity of the magnetic structure for $\text{Yb}(\text{Ni}_{1-x}\text{Cu}_x)_3\text{Al}_9$ ($x \leq 0.06$) [9]. Furthermore, formation of the chiral-soliton lattice is clearly observed in the magnetic field perpendicular to the helical axis. These results indicate that the asymmetric spin interaction really arises in the structure of YbNi_3Al_9 .

I will present the chiral magnetism and the Kondo-lattice properties of YbNi_3Al_9 and related compounds. I will also mention about the improvement on the flux method.

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Extremely large magnetoresistance and the complete determination of the Fermi surface topology in the semimetal ScSb

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Rare-earth monopnictides RX (where R = rare-earth, X = Sb, Bi) with the rocksalt-type structure have drawn attention recently because of their extremely large magnetoresistance (XMR) behaviour. An important member of this family is LaBi, which exhibits intriguing magneto-transport properties: a large non-saturating MR, a field-induced upturn followed by a plateau at low temperature in resistivity-temperature curve [1]. These properties have been attributed to the existence of surface states and the band inversion, or orbital texturing near the X-point in the Brillouin zone. However, similar transport properties have also been observed in other iso-structure compounds such as LaSb [1,2] and YSb [3], which have been proposed to be topologically trivial.

In this work, we present MR and Shubnikov-de Hass (SdH) oscillations of single crystalline ScSb, the smallest member in the RSb series. From our DFT calculations, the Fermi surface topology of ScSb is similar to that of LaBi and YSb but with two key differences. First, there is an additional hole pocket centred at Γ -point. Second, there is a large energy gap between the band formed by Sc d states and Sb p states near the X -point, which rule out the existence of band inversion.

The MR of ScSb shows non-saturating and nearly quadratic behaviour. Our angular dependent SdH oscillations detect *all* bulk Fermi pockets predicted by DFT calculations, including the additional hole pocket at Γ -point, and the electron pocket which was predicted in RSb compounds but not observed experimentally. Thus, ScSb can serve as a reference compound in which the bulk Fermi surface topology is fully determined to distill the essential ingredients responsible for the magneto-transport properties in RSb . Our study shows that the XMR behaviour can be attributed to the nearly perfect compensation of electron and hole carriers, indicating its semi-classical origin.

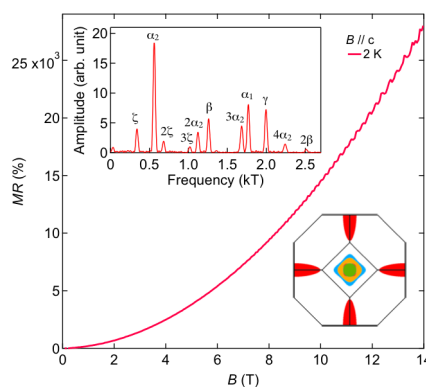


Fig. 1. Magnetoresistance of ScSb at 2 K, magnetic field $B \parallel c$ -axis. Insets show the corresponding FFT of SdH oscillation and Fermi surface.

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NMR studies of the incommensurate helical antiferromagnet EuCo₂X₂ (X = As, P)

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The Eu-based intermetallic compound EuCo₂X₂ (X = As, P) crystallizes in the ThCr₂Si₂-type structure (Space group: No. 139, *I4/mmm*, *D*¹⁷_{4h}). The Eu ion has the divalent state, and orders antiferromagnetically below the Néel temperature $T_N = 47$ K for EuCo₂As₂ [1, 2] and $T_N = 66.5$ K for EuCo₂P₂ [3-6], respectively. The antiferromagnetic (AFM) structure below T_N was reported to be helical from the neutron diffraction (ND) study [2, 3]. The Eu ordered moments are aligned ferromagnetically in the *ab*-plane with the helix axis along the *c*-axis [2, 3]. The magnetic structure is usually determined by using ND measurements. The AFM propagation vector *k* of the incommensurate helical state in EuCo₂X₂ (X = As, P) was successfully determined by using nuclear magnetic resonance (NMR) [7, 8].

In this study, we investigate magnetic properties of EuCo₂X₂ (X = As, P) from microscopic point of view by using NMR technique. NMR measurement is useful for determination of the magnetic structure in AFMs such as high-*T_c* cuprates and Fe-based superconductors.

In the AFM state below T_N , we succeeded in observing ¹⁵³Eu, ⁵⁹Co, ⁷⁵As and ³¹P NMR signal. The external magnetic field dependence of ¹⁵³Eu, ⁷⁵As, ³¹P NMR spectra for single-crystalline EuCo₂As₂ and EuCo₂P₂ clearly evidenced the incommensurate helical AFM structure. We determined the AFM propagation vector characterizing the incommensurate helical AFM state by ⁵⁹Co NMR at zero magnetic field for each compound.

The research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358. Part of the work was supported by the Japan Society for the Promotion of Science KAKENHI: J-Physics (Grant Nos. JP15K21732, JP15H05885, and JP16H01078). N. H. also thanks the KAKENHI: J-Physics for financial support to be a visiting scholar at the Ames laboratory.

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Structural, Magnetic and Electrical Transport Properties of Single Crystalline CeTAs_2 ($T = \text{Cu, Ag}$)

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The Ce-based compounds are known to be very attractive due to their exotic physical properties that arise from the competition between the RKKY interaction and Kondo effect. This competition leads to the Kondo behavior, heavy fermionic behavior, unconventional superconductivity, non-Fermi liquid behavior etc. This diverse nature in Ce-based compounds arises due to the proximity of $4f$ level to the Fermi energy. Recently, Ce based compounds CeTAs_2 ($T = \text{Cu, Ag}$) have drawn much attention due to the interesting structural and unusual physical properties [1-2]. The single crystals of CeTAs_2 ($T = \text{Cu, Ag}$) have been prepared by self-flux method. CeCuAs_2 is found to crystallize in HfCuSi_2 type non-centrosymmetric tetragonal crystal structure, with a space group $P4/nmm$ (No. 129), whereas CeAgAs_2 does not crystallize exactly in HfCuSi_2 type structure, instead it crystallizes in the four-fold superstructure with a space group $Pmca$ (No. 57). Laue diffraction pattern, transmission electron microscopy (TEM) analyses and powder X-ray diffraction data of the prepared CeCuAs_2 crystal reveal the HfCuSi_2 -type tetragonal structure, whereas the same analyses depict the orthorhombic crystal structure for the CeAgAs_2 single crystal. The superstructure nature in CeAgAs_2 can be clearly observed in the TEM pattern as shown in Fig. 1(b), which contrasts with the pattern obtained for CeCuAs_2 which exhibits HfCuSi_2 type tetragonal structure [Fig. 1(a)]. CeCuAs_2 does not order magnetically down to mK temperature, whereas CeAgAs_2 orders antiferromagnetically at ~ 6 K (T_{N1}) and 4.9 K (T_{N2}). Temperature and field variation of dc magnetization data of CeTAs_2 ($T = \text{Cu, Ag}$) when magnetic field has been applied along two principal crystallographic directions reveal that the basal plane (ab -plane) is the easy axis of magnetization. Temperature variation of paramagnetic susceptibility data along $[001]$ direction exhibit a broad hump indicating intermediate valence fluctuation in CeAgAs_2 . Electrical resistivity data of CeCuAs_2 reveal a logarithmic temperature dependence thereby indicating the presence of Kondo effect. A negative value of temperature coefficient of electrical resistivity has been observed over the whole measured temperature range indicating a behavior similar to a Kondo semiconductor. CeAgAs_2 also exhibits a logarithmic temperature dependent electrical resistivity above magnetic ordering which reveals a Kondo behavior.

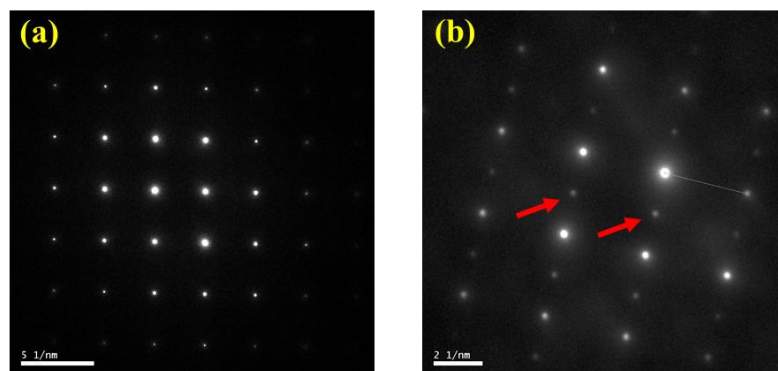


Fig. 1. (a) TEM image of CeCuAs_2 and (b) TEM image of CeAgAs_2 , red arrow lines indicate the superstructure.

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Quantum Oscillations in $\text{PrT}_2\text{Cd}_{20}$ ($T = \text{Ni, Pd}$) Compounds

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The RT_2X_{20} “cage” compounds, where R is a lanthanide or actinide ion and T is a transition metal ion, exhibit novel types of correlated electron phenomena, which are associated with the hybridization of localized f and d electron states with ligand states of the surrounding ions of the cages within which the R and T ions reside. Our laboratory discovered a new class of these compounds, with $X = \text{Cd}$ [1]. Additionally, $R = \text{Pr}$ compounds are of considerable interest due to widespread quadrupolar ordering and compounds exhibiting superconductivity [2][3] and the possibility of a multi-channel Kondo effect [4] as well as ferrohastatic order [5].

Previous study of $\text{PrNi}_2\text{Cd}_{20}$ and $\text{PrPd}_2\text{Cd}_{20}$ compounds has been inconclusive as to their ground states [6]. In this study, we examined their Fermi surfaces using quantum oscillations. Using pulsed fields up to 60 T at the Los Alamos National High Magnetic Field Laboratory, we performed Shubnikov de Haas (SdH) measurements, using a Proximity Detector Oscillator (PDO) method, on crystals of these compounds, as a function of angle using a rotator [7]. We were able to compare our results to theoretical calculations of $\text{LaNi}_2\text{Cd}_{20}$, which helped to further clarify the interpretation of the SdH data. Figures 1, 2, and 3 show the experimental results for $\text{PrNi}_2\text{Cd}_{20}$, $\text{PrPd}_2\text{Cd}_{20}$, and the theoretical results for $\text{LaNi}_2\text{Cd}_{20}$, respectively.

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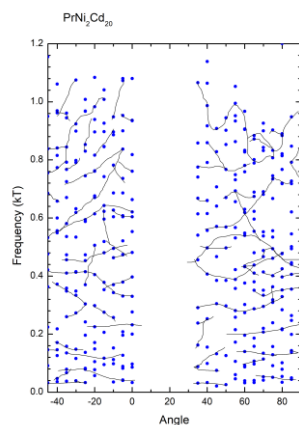


Fig. 1. SdH oscillation Fourier transform peaks vs. angle for $\text{PrNi}_2\text{Cd}_{20}$. The lines are guides for the eye.

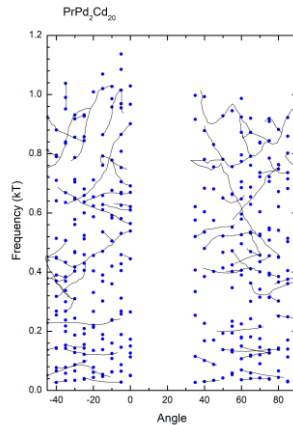


Fig. 2. SdH oscillation Fourier transform peaks vs. angle for $\text{PrPd}_2\text{Cd}_{20}$. The lines are guides for the eye.

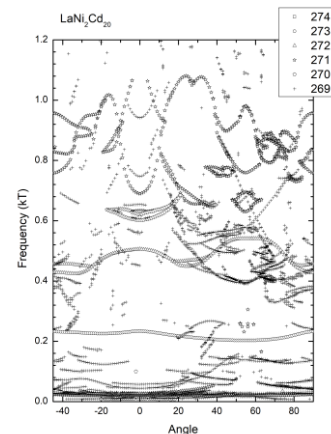


Fig. 3. Theoretical SdH oscillation Fourier transform peaks vs. angle for $\text{LaNi}_2\text{Cd}_{20}$

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Magnetic ordering in single-crystalline CeAgAs₂

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CeAgAs₂ crystallizes with an orthorhombic unit cell (s.g. *Pmca*), a derivative of the tetragonal HfCuSi₂-type (s.g. *P4/nmm*). Its characteristic feature is the presence of cis-trans chains of As atoms, the formation of which is a driving force for the symmetry breaking in the parent tetragonal structure [1,2]. The orthorhombic deformation is however fairly small and hence the compound shows almost isotropic physical behavior within the tetragonal plane of the parent unit cell [3]. Thus, measurements of single crystals of the compound were carried out along main directions of the parent unit cell.

CeAgAs₂ orders antiferromagnetically at the Néel temperature of 4.9 K, as revealed from low-temperature magnetization, specific heat and electrical resistivity data. The compound shows distinct magnetic anisotropy with easy magnetization direction perpendicular to the *c* axis of the parent tetragonal unit cell. Furthermore, clear metamagnetic transition occurs in a field of 0.5 T oriented within the easy magnetization direction. Observations of negative paramagnetic Curie temperature, moderately enhanced electronic contribution to the specific heat as well as characteristic temperature dependencies of the resistivity and the transverse magnetoresistance hint at the presence of Kondo interactions in the compound investigated.

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Superconducting properties and anisotropic superconducting gap of CeIr₃ single crystal

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The exploration of superconducting properties in Ce-based systems has been one of the most intriguing subjects in the research field of unconventional superconductivity[1]. In this study, we aim to elucidate superconducting properties of CeIr₃, which crystallizes in the PuNi₃-type rhombohedral structure with the space group R-3m (#166, D_{3d}^5). There are two crystallographically non-equivalent Ce sites (Ce1 and Ce2) and three Ir sites, as shown in Fig. 1. The superconductivity with a transition temperature of 3.34 K in CeIr₃ was first reported by Geballe *et al.* in the 1960s[2]. Previous studies were performed at zero field using polycrystalline samples, and the thermodynamic bulk properties have not been reported so far.

Single crystals of CeIr₃ were grown by the Czochralski method in a tetra arc furnace under an argon atmosphere. CeIr₃ has been reported to be an incongruent melting compounds. We tried the Czochralski method using starting materials with an off-stoichiometric ratio and succeeded in growing single crystals. We obtained single crystals with dimensions of approximately $1.5 \times 1.0 \times 1.0 \text{ mm}^3$, as shown in the inset of Fig. 2. The superconducting properties of CeIr₃ single crystal are investigated by electrical resistivity, specific heat, and magnetization. Figure 2 shows the temperature dependence of specific heat in the form of C/T and exhibits a clear jump corresponding to the superconducting transition at 3 K, indicating bulk superconductivity in CeIr₃. We will discuss the superconducting properties and the superconducting gap structure of CeIr₃ single crystal in the presentation.

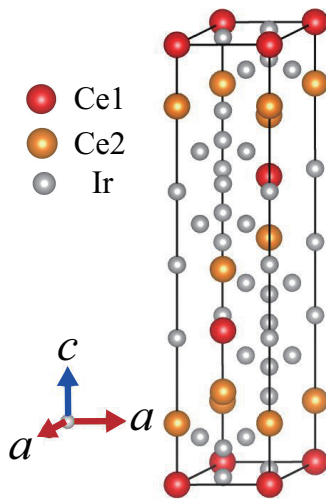


Fig. 1. Rhombohedral crystal structure of CeIr₃.

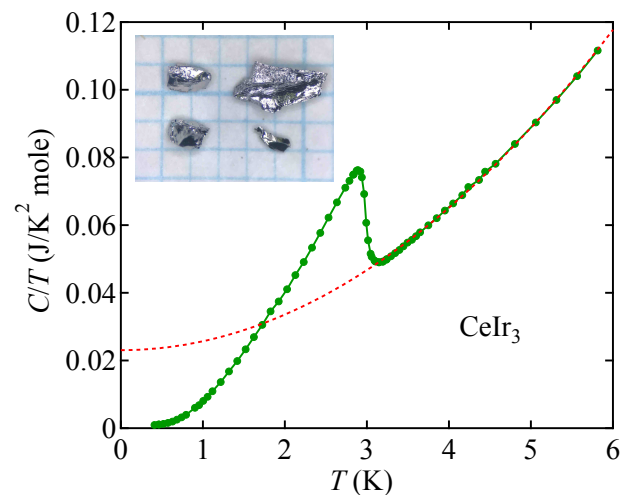


Fig. 2. Temperature dependence of the electronic specific heat of CeIr₃ single crystal. The inset shows pictures of CeIr₃ single crystal.

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A new way to control magnetism in Y-type hexaferrite

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Origins of the Magnetoelectric (ME) effect can be classified into three parts, magnetic hedgehog H , quadrupole Q and toroidal (dipole) moment T [1]. These multipoles appear in free energy in the form of $H(\mathbf{E} \cdot \mathbf{B})$, $Q_{ij}(E^i B^j + E^j B^i)$, and $T \cdot (\mathbf{E} \times \mathbf{B})$ with electric field $\mathbf{E}$ and magnetic field $\mathbf{B}$. Accordingly, the toroidal moment contributes to the electric polarization $\mathbf{P} \propto \mathbf{T} \times \mathbf{B}$ (magnetization $\mathbf{M} \propto \mathbf{T} \times \mathbf{B}$) induced by $\mathbf{B}$ ($\mathbf{E}$).

Y-type hexaferrites belong to the space group $R\bar{3}m$ (Fig. 1(a)) and show ME effect, where the low magnetic field $\mathbf{B}$ perpendicular to the c -axis induces the electric polarization $\mathbf{P}_{ab}$ normal to both $\mathbf{B}$ and the c -axis [2]. Its origin is accounted for by the spin-current mechanism [3] in the double-fan (transverse conical) magnetic structure (Fig. 1(b)) [4] and the structure has the magnetic toroidal moment. Here, there are two types of helicities in the cycloidal components of this structure and they induce electric polarizations opposite to each other. To align the helicity and following electric polarization, electric field normal to both magnetic field and the c -axis has been employed since only $\mathbf{P}_{ab}$ has been focused so far.

In addition to $\mathbf{P}_{ab}$, we experimentally verified that the electric polarization $\mathbf{P}_c$ along the c -axis is induced in the double-fan phase of $\text{BaSrCo}_2\text{Fe}_{11.1}\text{Al}_{0.9}\text{O}_{22}$. Figure 2 shows the displacement current $I_{\parallel c}$ along the c -axis under the rotation of in-plane magnetic field of 1.6 T at 100 K and at the speed of 0.58 degree/s. It is understood that there is electric current with the period of 120°. Its integration, in other words, changes in the electric polarization is also depicted in Fig. 2. This is in accordance with group-theoretical considerations, which predicts the coupling between the direction of $\mathbf{P}_c$ and the magnetic helicity. Therefore, this result may be employed to electrically control the magnetization, magnetic helicity and magnetic toroidal moment in Y-type hexaferrites. In this presentation, we discuss on the mechanism of this ME effect.

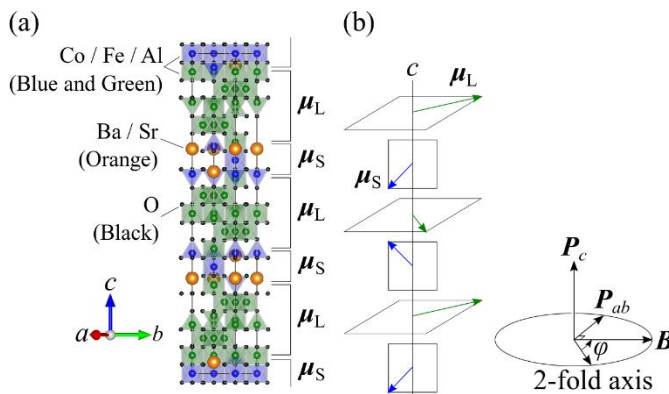


Fig. 1: (a)Crystal and (b)double-fan magnetic structure of Y-type hexaferrite $\text{BaSrCo}_2\text{Fe}_{11.1}\text{Al}_{0.9}\text{O}_{22}$

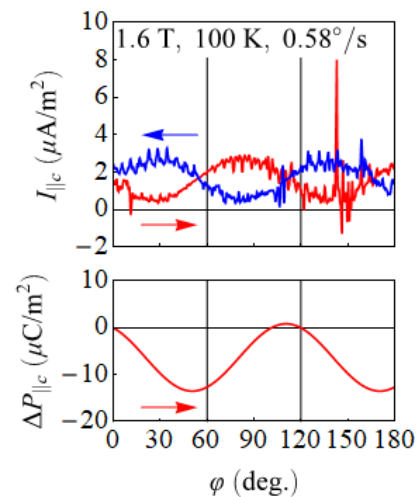


Fig. 2: Displacement current $I_{\parallel c}$ with respect to the rotation of in-plane magnetic field (1.6 T) at 100 K at the rate of 0.58°/s. Electric current with the period of 120° is integrated to obtain electric polarization.

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NMR evidence for anomalous magnetic ground state in field-insensitive Heavy Fermion System $\text{SmTa}_2\text{Al}_{20}$

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Recently, Yamada and co-workers reported that $\text{SmTa}_2\text{Al}_{20}$ shows field-insensitive magnetic transition and HF state [1]. This material exhibits Kondo-like $-\log T$ dependence of resistivity and the suppressed effective magnetic moment of $0.09\mu_B/\text{Sm}$ in the paramagnetic state, indicating strong c-f hybridization. λ -type specific heat peak was observed at around a phase transition temperature T_x , which is expected to be of an AFM type according to nuclear specific heat analyses. Below T_x , the 4f electron specific heat divided by temperature, C_{4f}/T , maintains a high value of $\sim 3 \text{ J}/(\text{molK}^2)$ down to 0.2 K. Such a largely enhanced specific heat coefficient below T_x suggests extremely heavy quasiparticles emerge in the AFM ordered state. The HF state as well as the magnetic phase transition is robust against magnetic fields. Such exotic behaviors are different from the conventional HF state. Although it has been discussed that either multipole or valence degrees of freedom plays important role in the field-insensitive behaviors in $\text{SmTa}_2\text{Al}_{20}$ [1,2,3] field-insensitive HF and a magnetic ordered states are still open issues. Thus, further investigations are required on $\text{SmTa}_2\text{Al}_{20}$.

In order to obtain more insight on 4f electron state of Sm ion in $\text{SmTa}_2\text{Al}_{20}$ from microscopic viewpoint, we have performed ^{27}Al -nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) studies on powdered single crystal $\text{SmTa}_2\text{Al}_{20}$.

Combining NMR/NQR spectra and numerical simulation, we successfully deconvoluted the NMR signals from the Al 16c, 48f, and 96g sites. The spectral broadenings of the NMR and NQR linewidth below T_x indicates the appearance of the internal field at the Al sites. From the spectral simulation, the ordered moment is estimated to be $\sim 0.07\text{--}0.25 \mu_B/\text{Sm}$. However, the zero-field NMR spectrum cannot be explained by the magnetic interaction solely, suggesting the existence of the electric interaction.

Details will be discussed at the J-Phys. Workshop.

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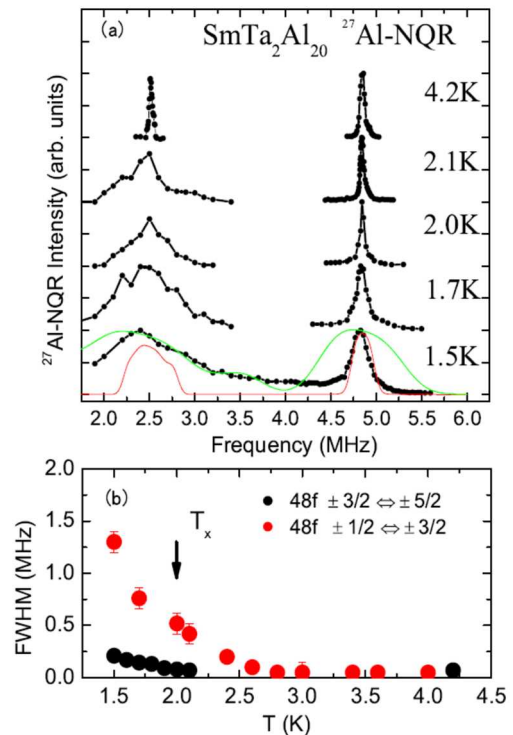


Fig. 1. (a) Temperature dependence of Al-NQR spectra for 48f site. (b) Temperature dependence of the linewidth.

Giant Phonon Softening and Enhancement of Superconductivity Induced by Copper/Phosphorus Doping of BaNi₂As₂

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The effects of copper and phosphorus substitution on the structural and superconducting phase transitions of BaNi₂As₂ were studied. BaNi₂As₂ crystallizes in the tetragonal ThCr₂As₂-type structure (space group I4/mmm, D_{4h}¹⁷, #139), and exhibits a first-order structural phase transition from its tetragonal phase to a triclinic phase (P-1, C_i¹, #2) at $T_s \sim 130$ K. In this triclinic phase, weak-coupling BCS superconductivity emerges at 0.7 K. We found that the structural phase transition was suppressed by P and Cu substitution for As and Ni of BaNi₂As₂, respectively, and superconducting transition temperature T_c increased abruptly from 0.7 K in the triclinic phase with less substitution to 2.5-3.3 K in the tetragonal phase with more substitution at $x = 0.067$ for BaNi₂(As_{1-x}P_x)₂ [1] and Cu substitution for Ni at $x = 0.16$ for Ba(Ni_{1-x}Cu_x)₂As₂ [2]. Specific-heat data suggested that doping-induced phonon softening was responsible for the enhanced superconducting transition temperature in the tetragonal phase, and the superconductivity in the tetragonal phase was a strong-coupling type, which was characterized by an enhanced specific heat jump at T_c . We also observed similar strong-coupling superconductivity at 3.5 K and the presence of soft phonon in BaPd₂As₂ with a tetragonal ThCr₂Si₂-type structure [3].

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Nonsaturating large magnetoresistance in high carrier density nonsymmorphic metal CrP

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The band structure of high carrier density metal CrP features an interesting crossing near the Y point of the Brillouin zone [1]. The crossing, which is protected by the nonsymmorphic symmetry of the space group, results in a hybrid, semi-Dirac-like energy-momentum dispersion relation near Y. The linear energy-momentum dispersion relation along Y- Γ is reminiscent of the observed bandstructure in several semimetallic extremely large magnetoresistance (XMR) materials [2].

We have measured the transverse magnetoresistance of CrP up to 14 T at temperatures as low as ~ 16 mK. Our data reveal a nonsaturating, quadratic magnetoresistance as well as the behaviour of the so-called ‘turn-on’ temperature in the temperature dependence of resistivity. Despite the difference in the magnitude of the magnetoresistance and the fact that CrP is not a semimetal, these features are qualitatively similar to the observations reported for XMR materials. Thus, the high-field electrical transport studies of CrP offer the prospect of identifying the possible origin of the nonsaturating, quadratic magnetoresistance observed in a wide range of metals.

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Low-energy phonon and *f* electronic state in the cage-structured compound $M\text{Be}_{13}$

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The beryllides $M\text{Be}_{13}$ (M = rare earths and actinides) crystallize in a NaZn_{13} -type cubic structure with $Fm\bar{3}c$ (No. 226, O_h^6) [1,2], where the M atoms in the $8a$ site are surrounded by 24 Be^{II} atoms in the $96i$ site, nearly forming an ideal snub cube, whereas the Be^{I} atoms in the $8b$ site are surrounded by 12 Be^{II} atoms, forming an icosahedron cage. Thus the $M\text{Be}_{13}$ systems can be categorized as a cage-structured compound. Among them, UBe_{13} is well known as the second heavy-fermion superconductor discovered by Ott *et al.* in 1983 [3]. Despite extensive studies over 30 years, the exact nature of the superconductivity and non-Fermi liquid state for UBe_{13} still remains unresolved. This puzzle may mainly stem from a lack of understanding of its $5f$ electronic and phonon properties. To obtain further insights into the novel features of UBe_{13} , it will be useful to reveal the common properties and differences in a series of isostructural $M\text{Be}_{13}$ compounds in detail.

In recent years, we have succeeded in growing single crystalline samples of $M\text{Be}_{13}$ by Al-flux method, and performed electrical resistivity, specific heat, magnetization, and X-ray diffraction measurements systematically, mainly for light-rare-earth $M\text{Be}_{13}$ compounds [4-6]. For the rare-earth $M\text{Be}_{13}$, it is confirmed that the c - f hybridization is negligibly weak, and their valence states are commonly pure trivalent, even for SmBe_{13} and EuBe_{13} , although only CeBe_{13} shows an intermediate valence state due to strong c - f hybridization [7]. The present study also suggests that the light-rare-earth $M\text{Be}_{13}$ compounds undergo a helical-magnetic ordering at low temperatures as well as heavy-rare-earth $M\text{Be}_{13}$ compounds and NpBe_{13} [8,9]. Furthermore, we obtained collateral evidence for the presence of the low-energy phonon modes common to the $M\text{Be}_{13}$ systems, which can be described well by a model assuming a conventional harmonic Einstein oscillation of the M atom with a characteristic temperature θ_E of ~ 160 K. This obtained value of θ_E is in a good agreement with the previous inelastic neutron scattering measurements for UBe_{13} and ThBe_{13} [10]. Interestingly, the obtained θ_E values in the present systems appear to be independent of either mass of the guest atom or guest free distance r_{gfd} in the snub cube, which is a characteristic feature not found in other cage-structured compounds.

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The ground states of the materials with similar zigzag structure to RuAs

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We will present the basic properties of some materials with similar zigzag structure to RuAs. RuAs has two successive metal-insulator transitions at $T_{M11} = 255$ K and $T_{M12} = 190$ K accompanied by structural transitions [1]. In the metallic phase, RuAs crystallizes in the orthorhombic structure with non-symmorphic $Pnma$ space group such that Ru zig-zag chain is formed (Fig.1). In the insulating ground state (below T_{M12}), X ray diffraction measurement for the single crystal has revealed that the superlattice formation into monoclinic $P2_1/c$ space group [3]. And the band calculations have suggested that the metallic phase possesses Fermi surface instability originating in the degenerated four-fold flat bands near the Fermi level. This degeneracy is protected by the non-symmorphic symmetry [2-3]. Therefore, a band Jahn-Teller effect, that is, a symmetry lowering from the non-symmorphic $Pnma$ structure is the key ingredient of metal-insulator transitions in RuAs. A coupling between the modulated crystal structure and the calculated number of the d electrons at each Ru site also proposes a formation of characteristic CDW.

Inspired by such a nature of the phase transition in RuAs, we synthesized polycrystalline samples of IrSi, NbCrP, TaFeGe and ZrFeGe which all crystalizes in orthorhombic $Pnma$ space group to search specific phenomena like RuAs in the similar non-symmorphic crystal systems. IrSi is an isostructural system of RuAs (Fig. 1), so that the similar Fermi surface instability to RuAs can be expected from band calculation [4]. NbCrP, TaFeGe and ZrFeGe have similar zigzag-chain of Nb (Ta, Zr) to RuAs (Fig. 2).

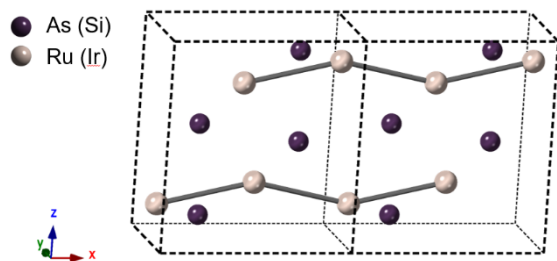


Fig. 1. Crystal structure of RuAs and IrSi.

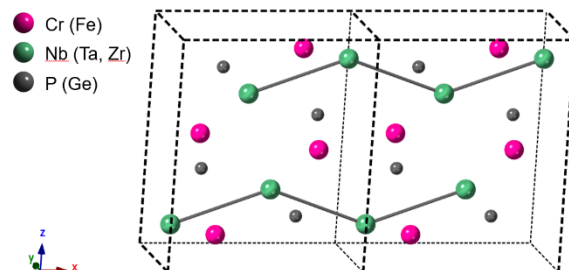


Fig. 2. Crystal structure of NbCrP, TaFeGe, and ZrFeGe.

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Magnetoelastic coupling in multiferroic $\text{CaBaCo}_4\text{O}_7$

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The coupling between electric, magnetic and elastic properties is expected in multiferroics. In multiferroics, the physical property is controlled by non-conjugate external field. In this research, we focused on the multiferroic $\text{CaBaCo}_4\text{O}_7$. $\text{CaBaCo}_4\text{O}_7$ is belonging to space group $Pbn2_1$, and an orthorhombic pyroelectric compound. The paramagnetic-ferrimagnetic transition is reported to occur at $T_c \sim 60$ K[1]. The easy axis of the ferrimagnetic moment is b axis. The electric polarization changes by 17 mC/m^2 at the magnetic transition[2]. Therefore the strong coupling between the lattice system and the magnetic property is expected in $\text{CaBaCo}_4\text{O}_7$.

As a first trial we investigated the elastic property of $\text{CaBaCo}_4\text{O}_7$ by an ultrasonic measurement. The temperature dependence of the elastic constant C_{11} and the magnetization M is shown in Fig. 1(a) and 1(b). C_{11} was measured using the longitudinal elastic wave of a frequency 33.488 MHz . The softening of C_{11} is observed at two temperatures. The relatively sharp softening is observed around the ferrimagnetic transition temperature T_c . On the other hand, the anomaly of the magnetization is hardly observed at the other softening temperature T^* . The anomaly of permittivity near T^* has also been reported[1]. The origin of the anomaly has not been clarified. In the poster presentation, we discuss the magnetic field dependence of C_{11} and the possible phase transition at T^* .

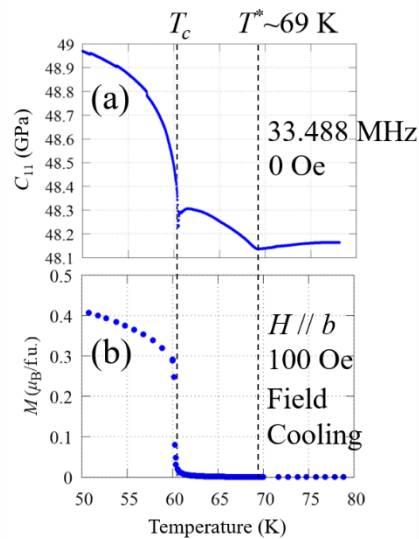


Fig. 1. Temperature dependence of (a) elastic constant C_{11} , (b) magnetization M .

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Antiferromagnetic quantum criticality and anomalous transversal resistivity in CeRh_2Si_2

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CeRh_2Si_2 with tetragonal structure has two antiferromagnetic transitions at $T_{N1} = 36$ K and $T_{N2} = 25$ K [1]. T_{N1} and T_{N2} decrease with increasing pressure and are suppressed to 0 at around $P_{c1} \sim 1$ and $P_{c2} \sim 0.5$ GPa, respectively. Around P_{c1} , the pressure-induced superconductivity [2] and the change of the Fermi surface topology [3] were reported. Recently, the existence of the current-induced magnetization was reported for the current along [100] direction at $T_{N2} < T < T_{N1}$ [4].

Figure 1 shows the temperature dependence of the longitudinal (ρ_{xx}) and transversal (ρ_{yx}) resistivity for the current along [100] (x -) direction on the plate-like sample perpendicular to the [001] (z -) direction. The temperature dependence of ρ_{xx} and ρ_{yx} are identical above T_{N1} . On the other hand, ρ_{yx} disagrees with ρ_{xx} below T_{N1} , which may be explained by the contribution of the anomalous Hall effect due to the current-induced magnetization along [001] direction or the anisotropic resistivity in the (001) plane. We will compare the both possibility for the origin of this anomalous transversal resistivity.

The pressure dependence of T_{N1} is well traced thanks to the anomaly in ρ_{yx} , although the anomaly in ρ_{xx} is smeared above ~ 1 GPa [2]. T_{N1} reaches zero at $P_{c1}' = 1.15$ GPa as shown in Fig. 2 (a), which is a bit but clearly higher pressure than $P_{c1} = 1.06$ GPa where the change of the Fermi surface related to the localized-itinerant transition of has observed. Figure 2 (b) shows that the onset of the superconductivity was widely observed from 0.9 to 1.3 GPa. On the other hand, zero resistivity was not confirmed below P_{c1} and the transition temperature of zero resistivity reaches the maximum between P_{c1} and P_{c1}' , and decreases above P_{c1}' . The low temperature resistivity deviates from the AT^2 dependence and A coefficient reaches the maximum between P_{c1} and P_{c1}' , which means that the antiferromagnetic fluctuations develop in the itinerant-antiferromagnetic state.

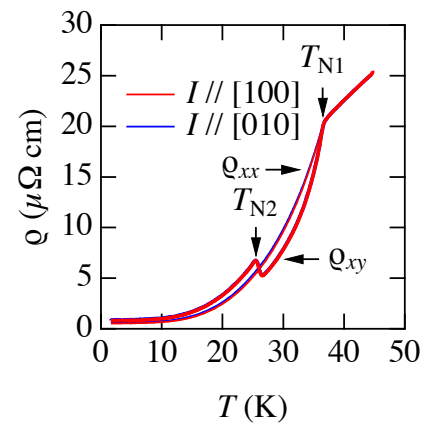


Fig. 1. Temperature dependence of longitudinal and transversal resistivity.

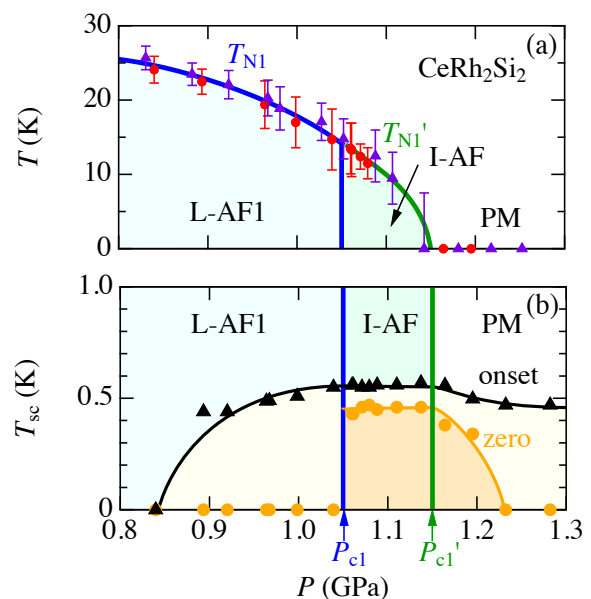


Fig. 2. Pressure dependence of T_{N1} (a) and T_{sc} of superconductivity (b).

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Antiferromagnetic transition in the doublet crystalline electric field ground state of $\text{NdT}_2\text{Zn}_{20}$ ($T = \text{Co, Rh}$)

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A family of caged compounds RT_2X_{20} (R : rare-earth, T : transition metal, X : Al, Zn, and Cd) crystallize in the cubic $\text{CeCr}_2\text{Al}_{20}$ -type structure [1]. The coordination for the R ion caged by 16 X atoms allows the $4f$ electrons under the cubic crystalline electric field (CEF) interact strongly with conduction electrons. For example, PrT_2X_{20} with a non-Kramers Pr^{3+} ions of the $4f^2$ configuration exhibit novel phenomena arising from the quadrupolar degrees of freedom [2]. Recently, NdT_2X_{20} with the Kramers Nd^{3+} ions of the $4f^3$ configuration have attracted much attention. In $\text{NdIr}_2\text{Zn}_{20}$, a Kramers Γ_6 doublet ground state undergoes an antiferromagnetic (AFM) transition at 0.65 K [3]. Theoretically, a two-channel Kondo effect has been predicted to occur in the Γ_6 doublet [4], which possibility deserves to be studied experimentally.

We have extended our work on NdIr_2Zn_2 to $\text{NdT}_2\text{Zn}_{20}$ with $T = \text{Co}$ and Rh . Because of the smaller lattice parameters than that for $T = \text{Ir}$, the interaction between the $4f$ and conduction electrons would be strengthened. The previous work for $T = \text{Co}$ suggested the CEF ground state to be the Γ_6 doublet but did not detect any phase transition down to 2 K [5]. To compare systematically the CEF level schemes and ground states among the compounds with $T = \text{Co}$, Rh , and Ir , we have grown single-crystalline samples and measured the magnetic, transport, and thermal properties down to 0.37 K.

The magnetic susceptibility $\chi(T)$ follows the Curie-Weiss law for 10–300 K with the effective magnetic moments of Nd^{3+} ions. The magnetic specific heat C_m data for both compounds show broad maxima at around 15 K, which are reproduced by a Schottky model with the Γ_6 doublet ground state and the excited Γ_8 quartet lying at around 40 K. On further cooling, the data of $C_m(T)$ for $T = \text{Co}$ and Rh exhibit sharp peaks at $T = 0.52$ K and 0.92 K, respectively, which coincide with drops observed in the electrical resistivity. With applying magnetic fields, the peak of C_m shifts to lower temperatures, indicative of an AFM transition. Interestingly, for the three compounds, the magnetic entropy S_m at T_N is only $0.5R\ln 2$ although the release of $R\ln 2$ is expected for the magnetic transition of the Γ_6 doublet. The reduction of S_m at T_N may result from short-range magnetic correlations and/or hybridization of the $4f$ electrons in Γ_6 doublet with conduction bands.

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⁷³Ge-NQR studies under pressure on magnetic fluctuations of ferromagnetic superconductor UGe₂

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UGe₂ crystallize in the orthorhombic ZrGa₂ type structure with the space group *Cmmm* and has a unique phase diagram. The superconducting (SC) state in UGe₂ arises inside the FM state and is observed in the pressure range of $P = 1$ to 1.5 GPa [1]. The magnetization shows the enhancement at T_X in the FM phase [2]. The transition between FM1 with smaller ordered moment and FM2 with larger one is a broad crossover at ambient pressure. However, the crossover region terminates by applying the pressure. The terminal point is called a critical point (CP), above which the FM1-FM2 transition is of a first order. The CP in UGe₂ is reported as $T_{CP} = 7$ K and $P_{CP} = 1.16$ GPa. The FM1-FM2 transition disappears above $P_X = 1.2$ GPa where the superconducting transition temperature is highest. This suggests the SC is involved in the FM1-FM2 transition in UGe₂.

The nuclear magnetic resonance (NMR) measurements have been performed in UCoGe which is one of the ferromagnetic superconductors [3, 4]. The measurements of the spin-lattice relaxation rate $1/T_1$ in UCoGe indicate that UCoGe possesses the Ising type magnetic fluctuation along the magnetic easy axis (*c*-axis) [3]. In addition, it was reported that the anisotropic fluctuation correlates closely with the SC [4]. On the other hand, the details of the magnetic fluctuations in UGe₂ have not been investigated. In order to clarify the anisotropy of the magnetic fluctuations in UGe₂, we performed the ⁷³Ge nuclear quadrupole resonance (NQR) measurement under pressure.

We measured T_1 and the spin-spin relaxation time T_2 to clarify the anisotropy of the magnetic fluctuations near T_X . At $P < P_X$, we found that the magnetic fluctuation along the *a* axis at the boundary between FM1 and FM2 is enhanced toward CP. Figure 1 shows the temperature dependences of $1/T_1T$ and $1/T_2$ in the FM2 phase at $P = 1.21$ and 1.32 GPa ($P > P_X$). $1/T_1T$ exhibits $T_1T \sim \text{const.}$ and has no anomaly. On the other hand, $1/T_2$ at both pressures shows complicated peak structure.

We will talk about the interpretation of the peaks of $1/T_2$. We also show the results of $1/T_1T$ and $1/T_2$ at $P = 1.44$ GPa.

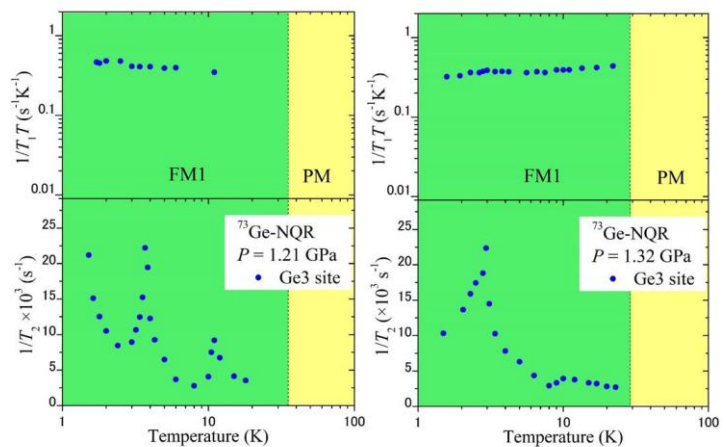


Figure 1. Temperature dependences of $1/T_1T$ and $1/T_2$ at $P = 1.21$ and 1.32 GPa.

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Effects of uniaxial stress on skyrmion-lattice host GaV_4Se_8

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GaV_4Se_8 and GaV_4S_8 , which are lacunar spinel selenides, are attracting attention as polar crystals hosting a Néel-type skyrmion lattice [1,2,3]. They belong to the noncentrosymmetric, nonpolar, achiral space group $F\bar{4}3m$ at the room temperature (Fig. 1). They undergo structural phase transition to the polar space group $R\bar{3}m$ at low temperatures, and when cooled further, magnetic orders occur. Since they elongate in one of $\langle 111 \rangle$ directions at the structural phase transition, four crystallographic domains can be formed. The existence of the crystallographic domains prevent us from investigating the anisotropy of the magnetic properties in detail.

In this research, we attempted to control crystallographic domains by applying uniaxial stress to GaV_4Se_8 single crystals. It is expected that when compressed in the $[111]$ direction in cubic setting, the domain elongating in the $[111]$ direction is minor. On the other hand, when compressed in the $[11\bar{2}]$ direction, the domain elongating in the $[111]$ direction may be dominant. Fig. 2 shows magnetization as a function of magnetic field in the $[111]$ direction under uniaxial stress at 5 K. The anomaly in the magnetization at 170 mT indicated by a triangular marker originates from the domains elongating in the $[1\bar{1}\bar{1}]$, $[\bar{1}1\bar{1}]$, and $[\bar{1}\bar{1}1]$ directions, whereas the others originate from the domain elongating in the $[111]$ direction. Comparing the magnitude of these anomalies, it is found that the domain elongating in the $[111]$ direction is about 50% of the whole sample without the uniaxial stress. The deviation from 25% may be attributed to the stress from substrate. By applying the uniaxial stress in the $[111]$ direction, the domain elongating in the $[111]$ direction decreases to about 10% of the whole sample. Moreover, the transition magnetic field from the skyrmion lattice phase to the forced ferromagnetic phase indicated by square markers changes by applying the uniaxial stress. It becomes smaller when compressed in the $[11\bar{2}]$ direction than that without the uniaxial stress. It suggests that compressive uniaxial stress in the direction parallel to the skyrmion lattice plane destabilizes the skyrmion lattice phase in GaV_4Se_8 . This is in contrast to the case in a chiral magnetic material MnSi in which it stabilizes the skyrmion lattice phase [4].

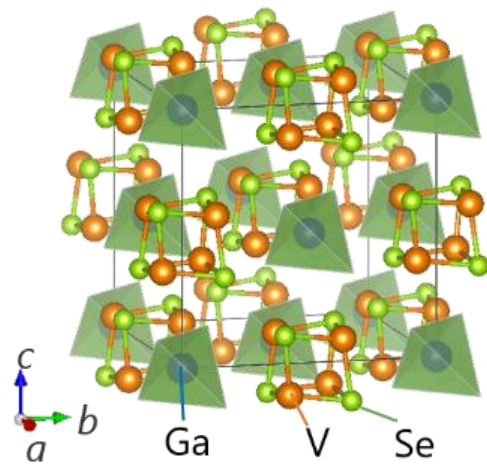


Fig. 1. Crystal structure of a lacunar spinel GaV_4Se_8 at the room temperature with the cubic space group $F\bar{4}3m$

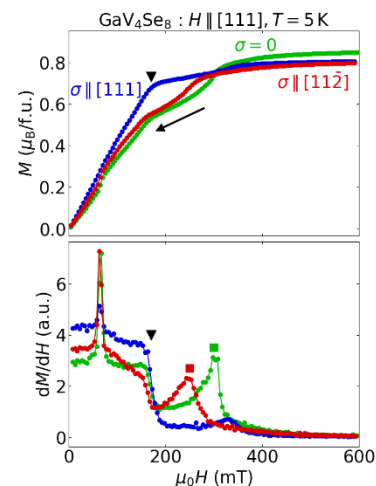


Fig. 2. Magnetization of GaV_4Se_8 as a function of magnetic field in the $[111]$ direction under uniaxial stress at 5 K

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Microscopic evidence for the dual Kondo temperatures in YbXCu_4 ($X = \text{Cu}, \text{Ag}$)

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Yb compounds YbXCu_4 ($X = \text{Cu}, \text{Ag}, \text{In}$), which crystallize in the cubic AuBe_5 -type (C15b) structure, exhibit a variety of physical properties depending on the X element: YbCuCu_4 is a heavy fermion with a large electronic specific heat coefficient $\gamma = 550 \text{ mJ/molK}^2$, YbAgCu_4 is a moderate heavy fermion with stronger valence fluctuations with $\gamma \sim 250 \text{ mJ/molK}^2$, and YbInCu_4 undergoes a first-order valence transition at 42 K. All these compounds show a localized (high temperature) - itinerant (low temperature) transitions with characteristic temperatures.

In this study, we have estimated local magnetic susceptibility from the Knight shift at the $16e$ and $4c$ sites of those compounds by nuclear magnetic resonance (NMR) measurement. This allows us to investigate the change of local charge distribution due to the localized-itinerant transition. For YbCuCu_4 and YbAgCu_4 , both bulk and local susceptibilities show two typical behaviors: the temperature dependence deviates from the Curie-Weiss law with decreasing temperature, followed by temperature-independent behavior with different constant values at low temperatures. It is obvious that the high-temperature and low-temperature states are attributed to localized and delocalized $4f$ electrons, respectively. One of the remarkable findings in the present study is that the local susceptibility at the two different sites in $X = \text{Cu}, \text{Ag}$ compounds reveals different characteristic temperatures below which the itinerant state evolves, indicating the existence of major and minor hybridizations between the $4f$ and conduction electrons.

We also show the results of YbInCu_4 in the presentation.

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NMR study on rattling properties of tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$

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“Rattling” is defined as large amplitude atomic oscillation of guest ions in anharmonic potentials inside the caged structure composed of host ions. Such anharmonic vibration suppresses the thermal conductivity and enhances thermoelectric performance. Tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$ has attracted attention because of its high thermoelectric performance and anomalous physical properties. Figure 1 shows the crystal structure of tetrahedrite. The crystal structure belongs to a body-centered-cubic (bcc) system with space group $I-43m$ (No. 217, $Z = 2$) at room temperature. There exist two Cu sites named as Cu(1) and Cu(2) and also two S sites named as S(1) and S(2). Cu(1) site is tetrahedrally surrounded by four S(1) atoms and Cu(2) site is trigonally surrounded by two S(1) atoms and one S(2) atom. It is reported that Cu(2) in tetrahedrite has large atomic displacement parameter (ADP) along the axis perpendicular to S_3 triangle plane similar to a rattling. However, tetrahedrite has no caged structure around the Cu(2) atom unlike other rattling system. Recently, this system has been regarded as noncaged rattling system [1].

Furthermore, tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$ exhibits metal-semiconductor transition (MST) at $T_{\text{MST}} = 85$ K. Crystal structure below T_{MST} and mechanism of transition are still unclear.

We studied rattling properties of $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$ by NMR.

Firstly, we studied on rattling dynamics from spin-lattice relaxation time T_1 . $1/T_1T$ increases in the temperature region above 170 K, which is different behaviour from usual metal and seems to be caused by rattling-related fluctuations.

Secondly, we studied on the MST from NMR spectra and T_1 . Cu-NMR spectrum changes abruptly below T_{MST} , which is the same result as previous work [2]. However, the line profile at $T < T_{\text{MST}}$ has not been well reproduced by exact-diagonalized spectrum simulation. Then we performed first-principles calculation to explain the NMR spectrum in the low-temperature region. We also measured T_1 . T_1 for both Cu sites shows drastic change below T_{MST} . Since T_1 reflects density of states at Fermi level, the present results evidence the change of the electronic state for each Cu sites.

We will report these results and discuss the relationship between the MST and the Cu rattling.

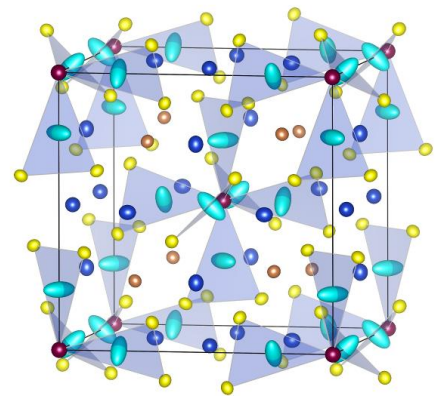


Fig. 1. Crystal structure and $S(1)_2S(2)$ plane of tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$. Ellipsoid reflects atomic displacement parameter [3].

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Nonmagnetic ground state doublet in a cubic Pr-based compound PrMgNi₄

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Recently, there has been considerable interest in cubic praseodymium-based compounds with nonmagnetic doublet ground states of $4f^2$ configuration, since they show a variety of phenomena originating from quadrupolar degrees of freedom. For example, PrPb₃ shows an antiferroquadrupole order at $T_Q = 0.4$ K [1], below which the electric quadrupoles are aligned with a sinusoidally modulated structure [2]. In addition, in PrT₂Zn₂₀ ($T = \text{Rh}$ and Ir) and PrT₂Al₂₀ ($T = \text{Ti}$ 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 [3].

In the present work, we have measured the electrical resistivity ρ , magnetic susceptibility χ , and specific heat of a Pr-based intermetallic compound PrMgNi₄ crystallizing in the cubic MgSnCu₄-type structure [4]. Since the Pr-ion sits at a cubic T_d site, the crystalline electric field (CEF) ground state could be the nonmagnetic doublet with the active quadrupoles. On cooling from 300K, $\chi(T)$ measured for a polycrystalline sample monotonically decreases. χ follows the Curie-Weiss law, where the effective magnetic moment μ_{eff} and the paramagnetic Curie temperature θ_p were estimated to be $\mu_{\text{eff}} = 3.53\mu_B/\text{Pr}$ and $\theta_p = -8.4$ K, respectively. With decreasing temperatures for $T < 5$ K, χ approaches a constant value, indicating a Van-Vleck paramagnetic state. The magnetic specific heat C_m shows a broad maximum at around 4 K, which can be well reproduced by a two-level model with the ground state doublet and an excited triplet with an energy gap of 12 K. Thereby, the CEF ground state of PrMgNi₄ is the nonmagnetic doublet.

For the nonmagnetic doublet ground state, it was expected that the active quadrupoles could play a role for lifting the entropy of the ground state doublet. However, no phase transition was observed down to the lowest temperature of 0.08 K. Instead, on cooling for $T < 0.4$ K, the data of C_m approach a constant value. This behavior can be reproduced by a random two-level model on the assumption that the splitting energy is uniformly distributed by 5 K. Taking the random two-level state with no phase transition into consideration, the degeneracy of the doublet ground state must be split by symmetry lowering of the Pr site due to possible atomic disorder. Since the Pr and Mg atoms sit at the sites with the same point group of T_d , they may be randomly exchanged, leading to a nonmagnetic singlet ground state with no active quadrupoles.

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Single crystal growth of uranium based heavy fermion compounds

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High quality single crystals of uranium based heavy fermion compounds were successfully grown using Czochralski method, flux method, and Bridgeman method. We present our technique and strategy to develop the new materials.

High quality single crystals of URhGe were grown by the Czochralski method in a tetra-arc furnace. Two annealing processes, that is high temperature annealing and low temperature annealing under ultra high vacuum for a long time, are quite important. After annealing process, single crystals were cut into many pieces, and the resistivity was measured down to 100mK using a homemade ADR combined with PPMS in order to check the sample quality and the superconducting transition. The obtained residual resistivity ratio (RRR) is up to 100, but it is quite limited in volume.

High quality single crystals of UCoGe were obtained from the similar processes without high temperature annealing. The RRR up to 100 was also obtained. Contrary to URhGe, the Ferromagnetic transition at 3K is not very clear even in the high quality single crystals. This is probably due to the proximity of ferromagnetic quantum criticality even at ambient pressure, or the fact that ferromagnetic transition is the weak first order as indicated by NMR experiments.

High quality single crystals of URu2Si2 were grown by the Czochralski method, and by the In-flux method. Clear Shubnikov-de Haas experiments were observed in both samples.

We also present other systems which were grown in Oarai and in Grenoble.

This work was done in collaboration with G. Bastien, B. Wu, A. Gourgout, G. Knebel, A. Pourret, D. Braithwaite, J. P. Brison, J. Flouquet, G. Lapertot,

Superconductivity in trilayer $(\text{PbSe})_n(\text{TiSe}_2)_m$ misfit compound

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Misfit crystalline compounds are layered materials with two different crystal structure alternatively stacked along the c -axis. There are various compositions known; for the chalcogenides, the components are limited to the combination of monochalcogenide of Sn, Pb, Sn, Bi, and rare earth elements and dichalcogenides of Ti, V, Cr, Mo, W, Nb, and Ta [1].

Superconductivity is observed in mostly Nb and Ta compounds thanks to the superconductivity of their original dichalcogenides. One exception is in Ti compound that composes $(\text{PbSe})_{1.16}(\text{TiSe}_2)_2$ at the transition temperature (T_c) of 2.3 K [2]. This is similar to the superconductivity observed in intercalated TiSe_2 such as Cu_xTiSe_2 [3]. The similarities lie in their anisotropic nature and the relation with the charge density waves that disappears as the superconductivity comes in.

There are only mono- or bilayer TMD compounds reported in the single crystals of misfit crystalline compounds so far. We have synthesized trilayer- TiSe_2 compound, $(\text{PbSe})_{1+\delta}(\text{TiSe}_2)_3$ for the first time and measured the superconductivity at 2.6 K as shown in Fig. 1. The crystal structure of stacking 3 TiSe_2 layers is confirmed by transmission electron microscopy. The X-ray diffraction along the c -axis also confirms the interlayer spacing of TiSe_2 as 3 layers compared to 2 layers. Similarity of intercalated compounds, the anisotropy of superconducting state is expected to be modified. In the presentation, we would discuss the difference between bilayer and trilayer compounds.

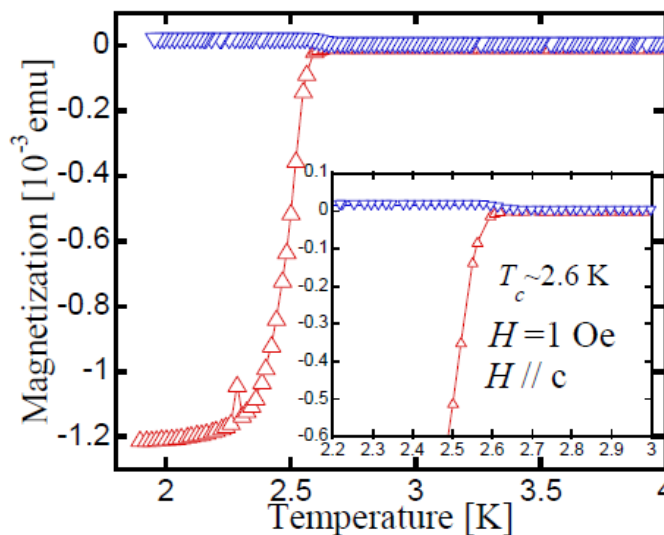


Fig. 1. Magnetization measurement on a single crystal of $(\text{PbSe})_{1.16}(\text{TiSe}_2)_3$. The large diamagnetic signal shows the superconductivity below 2.6 K.

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Spin-Glass Behavior in the Flux-Grown Single Crystal EuCu_2Si_2

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EuCu_2Si_2 crystallizes in the ThCr_2Si_2 -type tetragonal crystal structure. In the previous studies on arc-melted polycrystals, the valence state of Eu ions was reported to be nearly trivalent and fluctuate below room temperature.[1-5] Recently, single crystals of EuCu_2Si_2 were grown by the In-flux method. Interestingly, the Eu ions in the flux-grown single crystals were divalent, and a spin-glass behaviour was observed at low temperatures.[6-8]

To clarify the ground state of EuCu_2Si_2 and to get more insight into the spin-glass behaviour, we grew single crystals both by the Bridgman method and In-flux method and studied their magnetic, thermal, and electronic properties. Figures 1(a) and 1(b) show the temperature dependences of magnetic susceptibility for single crystalline EuCu_2Si_2 grown by the Bridgman method and In-flux method, respectively. The magnetic susceptibility for EuCu_2Si_2 (Bridgman) in Fig. 1(a) is very small, reflecting the nearly trivalence state of the Eu ions. On the other hand, the magnetic susceptibility for EuCu_2Si_2 (In-flux) follows the Curie-Weiss law at high temperatures, as indicated by solid lines in Fig. 1(b). In addition, when we measured the magnetic susceptibility at 0.01 T under field-cooled (FC) and zero-field cooled (ZFC) conditions, a large hysteresis between FC and ZFC conditions was observed for $H \parallel [001]$ below $T_{\text{SG}}=11$ K, as shown in the inset of Fig. 1(b). We will also present the results of crystallographic analyses of these single crystalline samples.

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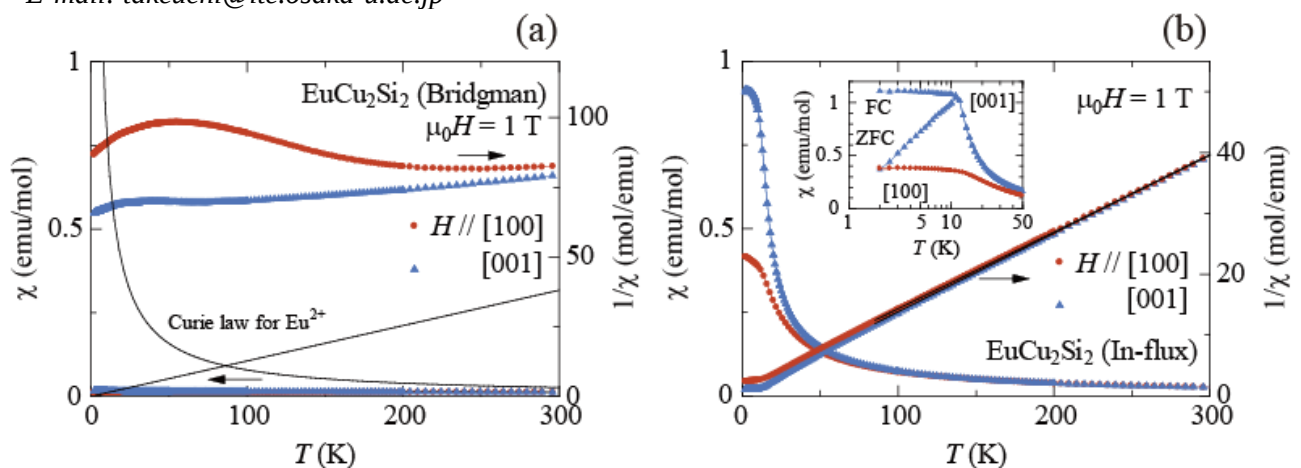


Fig. 1 Temperature dependences of magnetic susceptibility at $\mu_0 H = 1$ T for single crystalline EuCu_2Si_2 grown by the (a) Bridgman method and (b) In-flux method. The inset of (b) shows the data at 0.01 T for In-flux sample, showing a spin-glass behavior.

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P25 Katsuhiro Suzuki

NMR studies on magnetic fluctuations at low temperatures in $\text{PrT}_2\text{Al}_{20}$ ($T = \text{Nb, Ta}$)

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Intermetallic compounds RT_2X_{20} ($R = \text{rare earth}$, $T = \text{transition metal}$, $X = \text{Al, Zn, Cd}$) attract much attention because of their various intriguing phenomena at low temperatures such as ultra-heavy fermion state [1], quadrupolar Kondo effect [2,3], and field insensitive heavy fermion state [4,5]. Among them, $\text{PrT}_2\text{X}_{20}$ ($X = \text{Al, Zn}$) system shows quadrupole order, unconventional superconductivity, and non-Fermi liquid behaviors [6]. The origin of these novel behaviors is considered to be strong c - f hybridizations between conduction electrons and the multipolar active Γ_3 non-Kramers doublet CEF ground state of $4f$ electrons. Thus, $\text{PrT}_2\text{X}_{20}$ is believed to be an ideal playground to study the quadrupolar Kondo effect because of the coexistence of strong hybridizations and the higher order multipoles.

The NFL behaviors such as an upward convex curvature in the electric resistivity ρ are observed below 5 K in $\text{PrT}_2\text{Al}_{20}$ ($T = \text{Nb, Ta}$) [7,8]. $\text{PrNb}_2\text{Al}_{20}$ shows no long-range ordering down to 0.075 K [9] and $\text{PrTa}_2\text{Al}_{20}$ shows an antiferroquadrupole ordering at 0.65 K [7]. The T -dependences of ρ between 1 and 5 K in these compounds are well described by the quadrupolar Kondo lattice model [10].

In the present study, we performed ^{27}Al - and ^{93}Nb -NMR spin-lattice relaxation rate, $1/T_1$, measurements in $\text{PrT}_2\text{Al}_{20}$ ($T = \text{Nb, Ta}$). Figure 1 shows the temperature and field dependence of ^{93}Nb -NMR $1/T_1T$ between 3 and 19 T. $1/T_1T$ increases with decreasing temperatures in both compounds. At low temperatures, $1/T_1T$ in $\text{PrNb}_2\text{Al}_{20}$ has a broad maximum around 1 K, though $1/T_1T$ in $\text{PrTa}_2\text{Al}_{20}$ shows a sharp peak around 1 K, which is possibly due to the quadrupole order.

We will discuss these contrasting behaviors of magnetic fluctuations in these compounds based on the localized $4f$ CEF model and compare the field dependence of $1/T_1$ with another non-Kramers system such as $\text{PrOs}_4\text{Sb}_{12}$.

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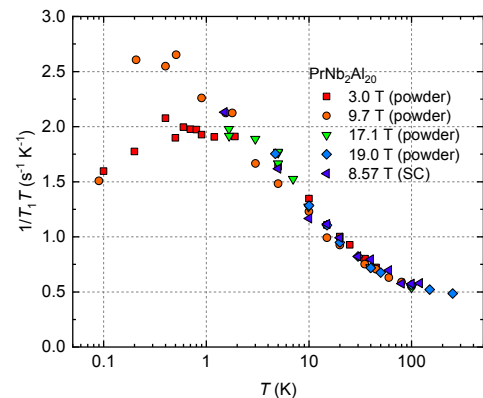


Fig. 1 T - and H -dependences of $\text{PrNb}_2\text{Al}_{20}$.

Magnetoelectric effect in a newly synthesized helical magnet $\text{Ni}_2\text{In}_{1-x}\text{A}_x\text{SbO}_6$ ($\text{A}_x = \text{Cr}_{0.1}, \text{Fe}_{0.05}$)

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In materials lacking inversion symmetry, noncollinear magnetic structures may arise from the competition between Dzyaloshinskii-Moriya interaction and Heisenberg exchange interaction. It is well known that the modulation of electric polarization by a magnetic field, a kind of magnetoelectric effect, can be observed in such noncentrosymmetric magnet. In addition, noncentrosymmetric helimagnets have recently been found to host topological magnetic ordering such as magnetic skyrmion. A long-period incommensurate helimagnet $\text{Ni}_2\text{InSbO}_6$ (NISO), which belongs to noncentrosymmetric chiral and polar space group $R3$, was first synthesized in 2013 [1, 2]. NISO has two crystallographically nonequivalent Ni^{2+} sites with $S = 1$ in the unit cell which are bonded antiferromagnetically and the helimagnetic modulation runs perpendicular to the c -axis. We have tried to realize ferrimagnetic-like helimagnetic configuration in magnetic ion doped NISO by means of the substitution of magnetic Fe^{3+} ($S = 5/2$) and Cr^{3+} ($S = 3/2$) for nonmagnetic In^{3+} . This strategy for material design works because the doped magnetic ions would be more strongly bonded to one of two nonequivalent Ni^{2+} ions.

In this study, we have succeeded in single-crystal growth of $\text{Ni}_2\text{In}_{1-x}\text{A}_x\text{SbO}_6$ ($\text{A}_x = \text{Fe}_{0.05}, \text{Cr}_{0.1}$). Neutron powder diffraction measurement confirmed an incommensurate magnetic modulation in zero field. Magnetization measurement revealed a magnetic phase transition in a magnetic field of about 3 T along the c axis, which is not seen in undoped NISO. Judging from the extrapolated value of magnetization to $\mu_0 H = 0$, the helical magnetic phase should change into a ferrimagnetic phase (Fig.1). We have found that this magnetic phase transition is accompanied by a change in electric polarization (Fig.1). As a result, the magnetoelectric response in low fields is enhanced by magnetic ion doping on NISO. We also propose magnetic phase diagram based on magnetization, electric polarization and dielectric constant measurement in high magnetic field (Fig.2).

In our presentation, we will introduce recent result of neutron diffraction experiment on single crystal of $\text{A}_x = \text{Cr}_{0.1}$ compound and discuss detailed magnetic structure of this system.

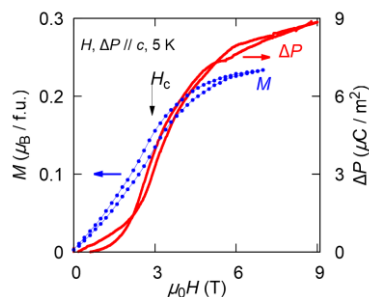


Fig. 1. M - H (blue) and ΔP - H (red) curves of $\text{Ni}_2\text{In}_{0.9}\text{Cr}_{0.1}\text{SbO}_6$. Electric polarization changes across the transition between helical and ferrimagnetic phase.

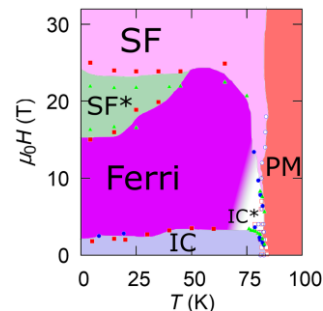


Fig. 2. Magnetic phase diagram of $\text{Ni}_2\text{In}_{0.9}\text{Cr}_{0.1}\text{SbO}_6$ under $H//c$. Collinear ferrimagnetic phase (violet region) is unique to doped system.

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Anisotropic Elastic Response in the Hidden Order Phase of URu₂Si₂ under High Magnetic Fields

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The elastic properties of URu₂Si₂ in the high-magnetic field region above 40 T, over a wide temperature range from 1.5 to 120 K, were systematically investigated by means of high-frequency ultrasonic measurements under the pulsed magnetic fields up to 68 T with pulse duration of about 150 ms at the Dresden High Magnetic Field Laboratory (HLD). Temperature dependence of the symmetry-breaking ultrasonic modes provides information on the bare $5f$ -electrons character with a broken of the unidentified electronic phase transition, so called ‘hidden order’, and weakened hybridization [1, 2]. We found contrast behaviours that appeared in three different transverse modes (as shown in Fig. 1); both Γ_4 (B_{2g}) and Γ_5 (E_g) symmetry modes of C_{66} and C_{44} show elastic softening which is enhanced above 30 T, respectively, while the characteristic softening of the Γ_3 (B_{1g}) symmetry mode $(C_{11}-C_{12})/2$ is suppressed in high magnetic fields. These results underscore the presence of a hybridization-driven Γ_3 (B_{1g}) lattice instability [3, 4] in the HO phase, and also in the strong c - f hybridization region at low-magnetic fields in URu₂Si₂. However, the enhanced softening of C_{44} and C_{66} for $H \parallel [001]$ at high magnetic fields cannot be fully explained by using existing CEF schemes applied to the quadrupolar susceptibility in a local $5f^2$ ($J = 4$) configuration. Thus, the origin of the softening in these transverse modes still remains an open question. Instead, we analysed the softening of $(C_{11} - C_{12})/2$ in low-magnetic field region by using the phenomenological theory for the band-Jahn-Teller effect. We will also report a recent progress on the ultrasonic measurements under static magnetic fields up to 28 T for $H \parallel [100]$ and $H \parallel [110]$.

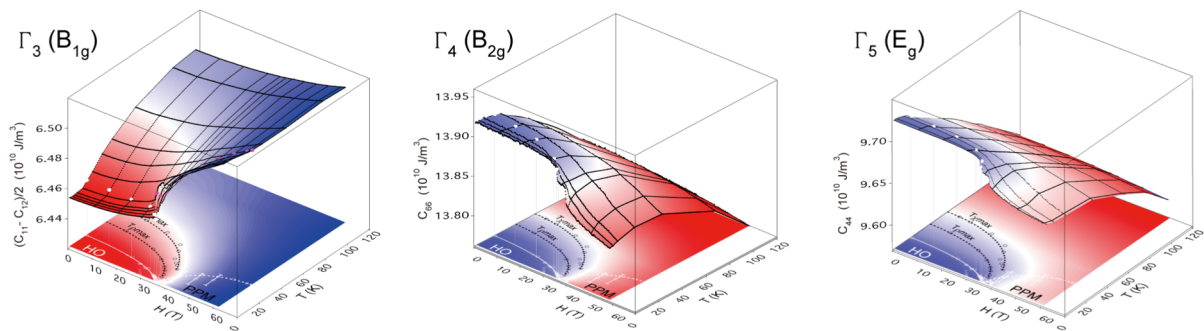


Fig. 1. Three-dimensional plots of the elastic constants of three transverse modes vs temperature and magnetic field aligned along the c axis of URu₂Si₂.

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Magnetization measurement of $\text{Ce}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Al}_{10}$ ($x < 0.05$) under Electric Current

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Toroidal moment $\mathbf{t}$ is one of the parameters that describe strength of the magnetoelectric coupling. According to a theoretical prediction by S. Hayami *et al.*, the toroidal moment can be active even in metallic systems, where the occurrence of exotic phenomena, such as magnetization induced by electric current, is expected [1]. Recently, we experimentally found the electric current-induced magnetization in an antiferromagnetic (AFM) ordered state of a metallic system UNi_4B [2]. However, the observed magnetoelectric effects in UNi_4B cannot be explained in part by the theoretical model. Further investigation in other metallic systems with the toroidal moment is required for full understanding of the magnetoelectric effect.

We now focus our attention on another AFM metallic system $\text{CeRu}_2\text{Al}_{10}$ and its Rh-doped system $\text{Ce}(\text{Ru}_{0.95}\text{Rh}_{0.05})_2\text{Al}_{10}$. They crystallize into $\text{YbFe}_2\text{Al}_{10}$ -type orthorhombic structure with Cmcm (D_{2h}^{17} , No. 63), where Ce ions align along c -axis forming zigzag structure, with no local inversion symmetry. Magnetic moments of the Ce ions order antiferromagnetically pointing to the c -axis at $T_N = 27$ K for $\text{CeRu}_2\text{Al}_{10}$, while the magnetic moments of $\text{Ce}(\text{Ru}_{0.95}\text{Rh}_{0.05})_2\text{Al}_{10}$ point to the a -axis with the AFM ordering below $T_N = 24$ K [3,4]. Both AFM orders have same propagation vector of $\mathbf{q} = (0, 1, 0)$. In the present study, dc M measurements under electric current for each setting of $\mathbf{I} \parallel a$ and c , and $\mathbf{H} \parallel a, b$, and c have been performed using large single crystalline samples provided by H. Tanida. Figure 1 shows the temperature dependence of M under $\mathbf{I}$ for $\text{CeRu}_2\text{Al}_{10}$. It is found that the additional M (ΔM) is induced by applying electric current below T_N for the three settings: $(\mathbf{I}, \mathbf{H}) \parallel (a, c)$, (c, a) , and (c, b) . Similar ΔM are also observed in $\text{Ce}(\text{Ru}_{0.95}\text{Rh}_{0.05})_2\text{Al}_{10}$. We are going to discuss about the cause of the observed magnetoelectric effects in these compounds based on the concept of odd-parity multipoles.

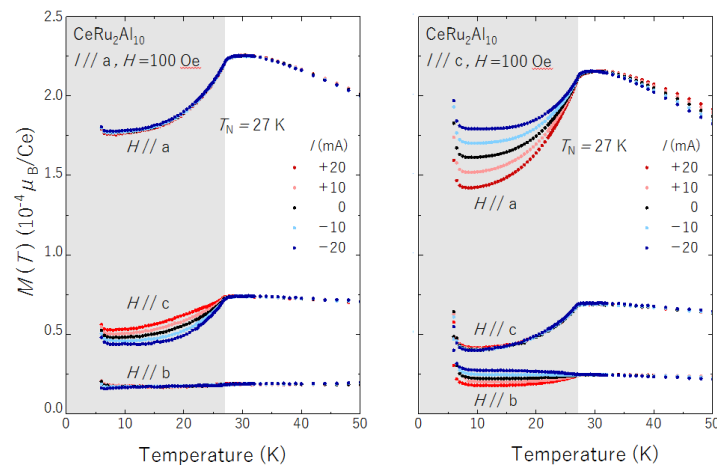


Fig. 1. Temperature dependence of M under electric current at $H = 100$ Oe for $\text{CeRu}_2\text{Al}_{10}$. (left) $\mathbf{I} \parallel a$, and $\mathbf{H} \parallel a, b$, and c ; (right) $\mathbf{I} \parallel c$, and $\mathbf{H} \parallel a, b$, and c .

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Single crystal growth and magnetic properties of non-centrosymmetric compound UIrSi₃

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A lack of inversion symmetry in the crystal structure gives rise to an antisymmetric spin-orbit interaction, which leads to the possibility of a superconducting state with an admixture of spin-triplet and spin-singlet pairs [1]. A huge upper critical field was observed in a non-centrosymmetric (NCS) heavy-fermion superconductor (HFSC) CeIrSi₃, which crystallizes with the BaNiSn₃-type tetragonal structure (*I4mm*), at the quantum critical point [2]. The magnetic ordering in these materials is usually antiferromagnetic with complex propagation vectors. We started to study the U compound as analogue to these Ce pressure-induced HFSCs. The U compound is not superconducting but has interesting magnetism.

UIrSi₃ orders antiferromagnetically below 42 K. After several attempts, we succeeded to grow a single crystal of UIrSi₃, for the first time, with a floating zone method (Fig.1) and measured its magnetization, specific heat, and electrical resistivity as functions of temperature, magnetic field, and pressure [3]. The results provide clear evidences of Ising-like antiferromagnetism below $T_N = 41.7$ K. Metamagnetic transition was observed at H_c ($\mu_0 H_c = 7.3$ T at 2 K) with the field along the *c*-axis. At low temperatures (< 28 K), it is a first-order transition and the asymmetry of hysteresis is attributed to gradual re-entry of the spin arrangement from the field polarized paramagnetic regime in fields above H_c to a complex ground state antiferromagnetic structure. H_c decreases with increasing temperature whereas the asymmetric hysteresis becomes rapidly reduced and vanishes at 28 K. At higher temperatures, it becomes second-order transition which is manifested as a cusp at H_c which terminates a pronounced upturn of the $M(H)$ dependence in lower fields. The point of change of the order of transition in the established H - T magnetic phase diagram, as shown in Fig.2, is tentatively suggested to be the tricritical point (at $T_{tc} = 28$ K and $\mu_0 H_{tc} = 5.8$ T) of UIrSi₃.

This work was supported by JSPS KAKENHI with Grant Nos. JP15K05156 and JP15KK0149.

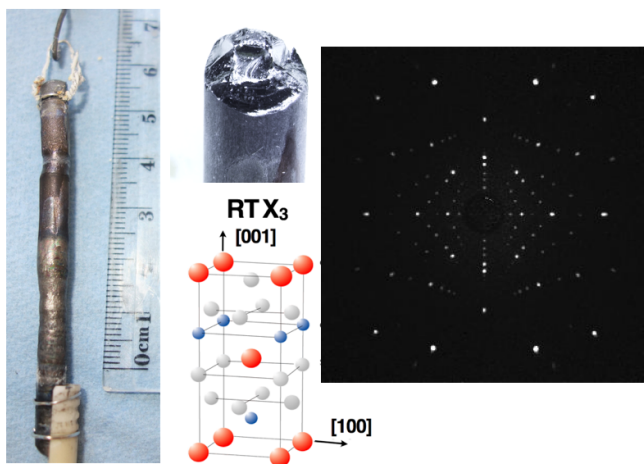


Fig. 1. Photograph, crystal structure and Lauegram of UIrSi₃.

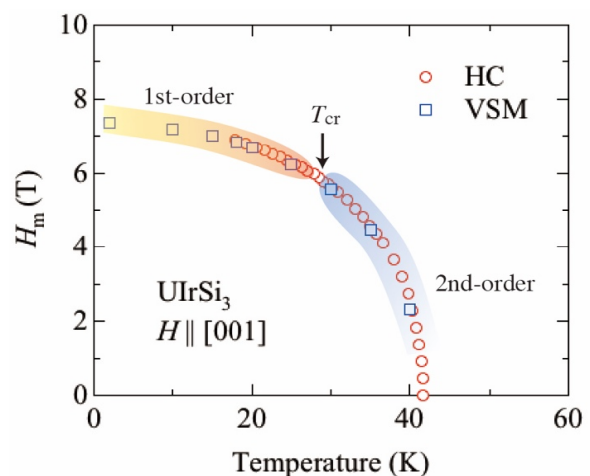


Fig. 2. H - T phase diagram of UIrSi₃ for $H \parallel [001]$.

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Magnetism throughout the UCoGe-URhGe-UIrGe system studied on alloy single crystals

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The ferromagnetic superconductors UCoGe and URhGe have the easy magnetization axis along the orthorhombic c -axis. Magnetization behavior in fields along b , however, plays important role in physics of these materials. URhGe at 2 K undergoes a first-order metamagnetic transition at $H_R = 12.5$ T whereas the $M(H)$ curve of UCoGe exhibits a broad S-shape around $H_m \approx 50$ T. The $\chi_b(T)$ curve of URhGe shows a sharp peak at $T_{\max} = 9.5$ K ($= T_C$) but a broad maximum around $T_{\max} = 37$ K ($\gg T_C = 3$ K) for UCoGe[1]. Similar feature – $T_{\max}$ was also detected in isostructural and isoelectronic antiferromagnet UIrGe ($T_N = 16.5$ K) along the identical axis[2, 3]. We prepared single crystals throughout UCo_{1-x}Rh_xGe and UIr_{1-x}Rh_xGe systems to establish a complex d_{U-U}/T (d_{U-U} is the shortest interatomic uranium distance and T is temperature) magnetic phase diagram. Our detailed study of single-crystals has revealed the evolution of the characteristic temperature ($T_{\max}$) and magnetic fields (H_R , H_m) as a function d_{U-U} parameter tuned by Co/Rh and Rh/Ir alloying. This recognized a characteristic maximum in magnetic susceptibility at temperature $T_{\max}$ along the b axis as an important parameter. Three magnetically ordered regions can be distinguished within this scope; first a ferromagnetic region between UCoGe and UCo_{0.7}Rh_{0.3}Ge with $T_C < T_{\max}$, second ferromagnetic region between UCo_{0.6}Rh_{0.4}Ge and UIr_{0.43}Rh_{0.57}Ge $T_C \approx T_{\max}$ and finally an antiferromagnetic region existing between UIr_{0.45}Rh_{0.55}Ge and UIrGe with $T_N < T_{\max}$ [4] (Fig. 1).

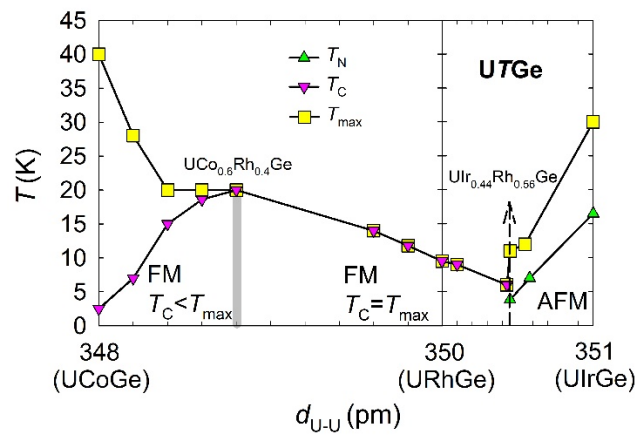


Fig. 1. d_{U-U}/T phase diagram of the UCoGe-URhGe-UIrGe system.

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Orbital Crossing on Split Fermi Surfaces in Noncentrosymmetric Yb_4Sb_3

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In noncentrosymmetric materials, spin-degenerated band splits into two bands due to antisymmetric spin orbit interaction (ASOI). Observation of such split Fermi surfaces by the parity-violation can unveil some concealed spin-related phenomena. For example, in heavy fermion system like CeRu_2Si_2 and CeB_6 , spin dependent mass enhancement was suspected to be realized [1,2]. However, there was no direct evidence for it since these Fermi surfaces are spin-degenerate due to their centrosymmetric crystal structures. This intriguing property was experimentally confirmed in noncentrosymmetric heavy fermion compound CeCoSi_3 [3]. Here, we report another example to reveal a new spin-related phenomenon exhibiting in a noncentrosymmetric intermediated-valence compound Yb_4Sb_3 via the de Haas-van Alphen (dHvA) effect. Theoretically expected electronic structure of Yb_4Sb_3 consists of pairs of split Fermi surfaces due to the ASOI [4,5]. We found multiple, more than two, dHvA signals from some of the pairs. We attribute them to the result of orbital crossing at degenerate points of the split Fermi surfaces, which is similar to the magnetic breakdown. There are two paths of carrier: one is intersecting and the other is passing-through the crossing (degenerate) point. In the latter path, the spin flip will take place at the crossing points. Strikingly, the probability of the intersecting is different in each Fermi-surface pair and field-direction. This result suggests that a novel transport property related to the spin structure characterized by the ASOI is realized in Yb_4Sb_3 .

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