PB01

Magnetic and Electrical Properties in $NpFe_4P_{12}$

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The recent discovery of superconductivity in $PuCoGa_5$ and $PuRhGa_5$ reveals that it is important to study the transuranium compounds including Pu and Np. Nevertheless it is very difficult to treat transuranium compounds experimentally because of the high radio activity and requirements of special equipment and technique.

We succeeded in growing single crystals of $NpFe_4P_{12}$ by the Sn-flux method and measured the electrical resistivity, magnetic susceptibility and magnetization. This is the first single crystal growth of transuranium filled-skutterudite compounds. The inset of Fig. 1 shows a photograph of $NpFe_4P_{12}$ single crystal. The flat surfaces corresponding to the (100), (110) and (111) planes, which were confirmed by the Laue pattern method, clearly appear. The lattice constant was determined as 7.7702(7) Å by the X-ray diffraction analysis.

We show in Fig. 1 the temperature dependence of resistivity for the current along $\langle 100 \rangle$ direction in $NpFe_4P_{12}$. The large value of resistivity indicates that the $5f^3$ configuration based on the $5f$-localized model is most likely applicable to $NpFe_4P_{12}$. The peculiar temperature dependence with nagative $d\rho/dT$ was observed between 30 K and 150 K, which is resemble to the results of the ferromagnet $NdFe_4P_{12}$.

Figure 2 shows the magnetization curves at 5 K for $H \parallel \langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$. The inset shows the temperature dependence of the magnetization at 1 kOe. $NpFe_4P_{12}$ orders ferromagnetically at 23 K. The ferromagnetic easy-axis was found to be $\langle 100 \rangle$ and the saturated moment is equal to 1.35 μ_B/Np.

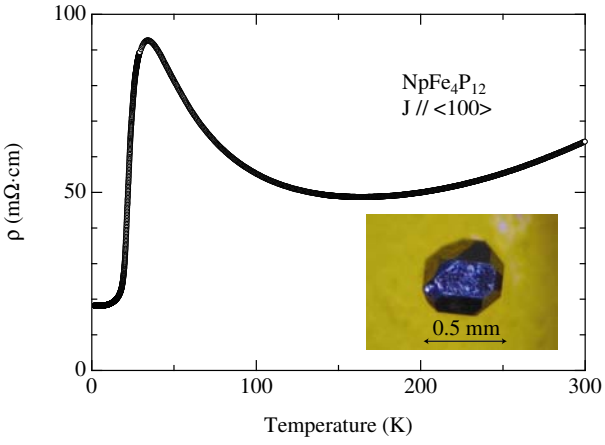

Figure 1: Temperature dependence of resistivity in $NpFe_4P_{12}$. The inset shows a photograph of $NpFe_4P_{12}$.

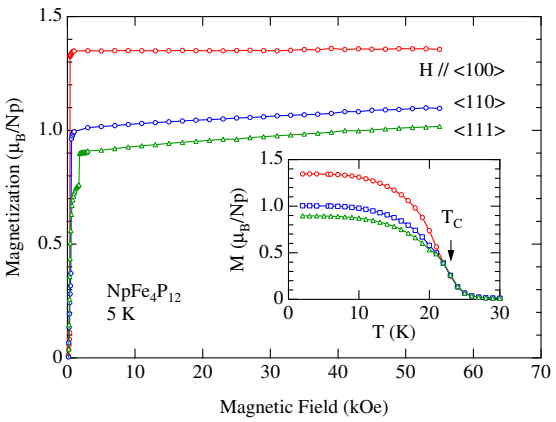

Figure 2: Magnetization curves at 5 K in $NpFe_4P_{12}$. The inset shows the temperature dependence of magnetization at 1 kOe.

PB02

NMR studies in U- and Np-based filled skutterudite compounds

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In the family of filled skutterudites, the only uranium- or transuranium-based compound successfully crystallized had been UFe_4P_{12}. There had been no other filled skutterudite which contains 5f electrons. Magnetization and resistivity measurements have indicated that UFe_4P_{12} is a ferromagnetic insulator with a Curie temperature $T_C = 3$ K [1-3]. Results from our recent ^{31}P-NMR studies of this compound have been reported in Ref.[4].

Recently, D.Aoki *et al.* have succeeded in growing single crystals of $NpFe_4P_{12}$ [5]. This new filled skutterudite compound is isostructural with UFe_4P_{12}, with the U replaced by Np. The lattice constant is found to be 7.7709 Å, which is the smallest value of all the filled skutterudite compounds. Magnetization measurements indicate the occurrence of ferromagnetic ordering with $T_C = 23$ K. On the other hand, the electrical resistivity shows a peculiar temperature dependence with a negative $d\rho/dT$ between 30 K and 150 K. In order to gain further insight into the electronic state of this new compound, we are now carrying out ^{31}P-NMR measurements using a single crystal.

[1] G. P. Meisner, M. S. Torikachvili, K.N.Yang, M. B. Maple and R. P. Guertin, J. Appl. Phys. **57** 3073 (1985).
[2] M. S. Torikachvili, C.Rossel, M.W. McElfresh, M.B.Maple, R.P. Guertin and G.P. Meisner, J. Magn. Magn. Matter. **54-57** 365 (1986).
[3] T. D. Matsuda, A.Galatanu, Y. Haga, S. Ikeda, E. Yamamoto, M. Hedo, Y. Uwatoko, T. Takeuchi, K. Sugiyama, K. Kindo, R. Settai and Y. Ōnuki, J. Phys. Soc. Jpn. **73** 9 (2004).
[4] Y.Tokunaga, T.D.Matsuda, H.Sakai, H.Kato, S.Kambe, R.E.Walstedt, Y. Haga, Y. Ōnuki, and H. Yasuoka, Physical Review B **71**, 045124 (2005).
[5] D.Aoki *et al.*, A01, A04 班合同研究会「充填スクッテルダイト化合物の試料育成の現状と展望」(室蘭工大).

PB03

Magnetic property of pressure–induced insulating–state in $PrFe_4P_{12}$

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$PrFe_4P_{12}$ exhibits an antiferroquadrupolar ordering at ambient pressure and a pressure–induced metal–insulator (M–I) transition above 2.4 GPa[1]. Recently we have carried out Hall effect and P-NMR measurements under high pressure. These results suggest that (1)a band–insulating state realizes (2)the M–I transition is of first–order (3)an internal field exists in the insulating phase. To make clear the magnetic property of the insulating phase, we carried out ac–susceptibility measurement under hydrostatic pressure and magnetization (M) measurement under uniaxial pressure. It is confirmed by our electrical resistivity measurement that the uniaxial pressure effect induces the M–I transition at lower pressure.

Figure 1(a) shows the temperature dependence of magnetization $M(T)$ at 0.1 T. M at 2.76 GPa is smaller than that at ambient pressure below 10 K and $M(T)$ has a broad maximum around 8 K. This result at 2.76 GPa almost corresponds to that of ac–susceptibility measurement at 3.6 GPa showing the M–I transition at 8 K. The magnetization process $M(H)$ at 2K is shown in Fig. 1(b). $M(H)$ at 2.76 GPa exhibits a metamagnetic behavior around 3 T. It is considered that the metamagnetic behavior is caused by the transition from insulator to metal. The value of M at zero field is zero, so that ferro or ferri magnetic ordered state could be excluded from the candidates for the magnetic ordered state of the insulating phase. An antiferromagnetic ordered state might realize in the insulating phase.

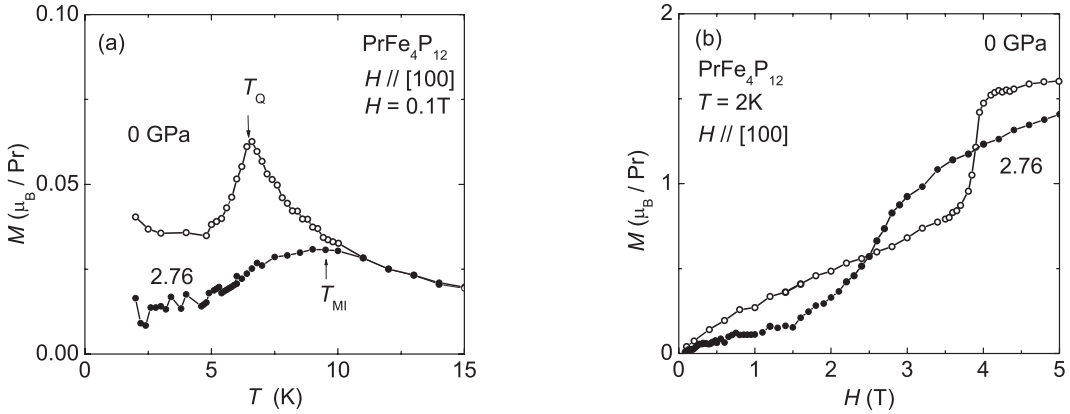

Figure 1: (a)T dependence of M at 0.1 T and (b) magnetization process at 2 K. The magnetic fields and the uniaxial pressure are applied parallel to [100].

[1] H. Hidaka *et al.*, Phys. Rev. B **71**, (2005) 073102.

PB04

Effect of pressure on the thermal expansion of $PrFe_4P_{12}$

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A high-pressure study displays a metal-insulator (M-I) transition above 2.4 GPa in $PrFe_4P_{12}$ which shows an antiferro-quadrupoler (AFQ) ordering below T_Q =6.5 K at ambient pressure[1]. We have studied the thermal expansion of $PrFe_4P_{12}$ under high pressure up to 3.0 GPa. Fig. 1 shows the temperature dependence of the linear thermal expansion ($\Delta L/L$) under pressure along a-axis. There is a small anomaly in $\Delta L/L(T)$ at T_Q= 6.5 K at ambient pressure. T_Q decreases with applying pressure at a rate of $dT_Q/dP \sim 0.8$ K/GPa. Above 2.8 GPa, a large anomaly is observed in $\Delta L/L(T)$ where M-I transition occurs.

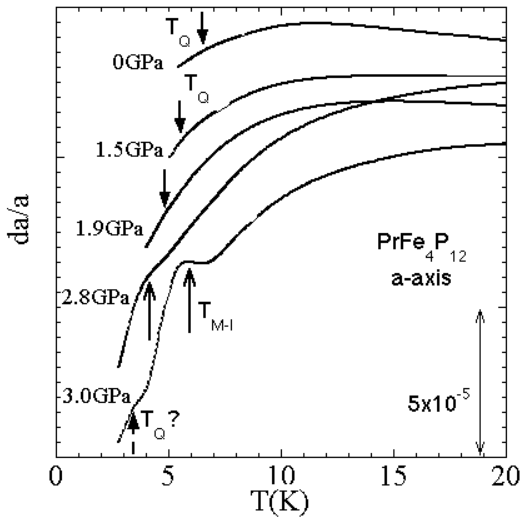

Figure 1: Thermal expansion of $PrFe_4P_{12}$ as a function of temperature under high pressure.

[1] H. Hidaka et al., Phys. Rev. B 71 (2005) 073102.

PB05

On the nature of the quadrupole-ordered phases of $PrFe_4P_{12}$: ^{31}P NMR study

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$PrFe_4P_{12}$ undergoes a phase transition into an antiferro-quadrupole (AFQ) ordered state below 6.5 K at zero field. The ordered phase is suppressed by applying magnetic field and there appears a non-ordered heavy-fermion state above about 4-7 T depending the direction of a field [1]. An interesting exception is the case of a field applied along [111], for which we observe non-fermi-liquid behavior and some ordered phase at very low temperatures and high fields [2]. In order to elucidate the nature of this high-field ordered phase, we measured ^{31}P NMR in $PrFe_4P_{12}$ using a single crystal.

Figure 1 shows temperature variation of the ^{31}P NMR spectrum under the [111] field. It is apparent that the lines exhibit neither splitting nor broadening across the transition temerature $T_B \approx 0.70$ K at 13 T. The frequency shifts of both the lines indicate no anomaly at T_B as well. These observations make a clear contrast with the case of the AFQ transition at low fields, for which the NMR lines split due to an appearance of the staggered magnetic field at the ^{31}P site [3]. On the other hand, we observed non-exponential recovery of the ^{31}P nuclear magnetization below T_B. This suggest either presence of several ^{31}P sites which cannot be disinguished by a measurement of the spectrum, or spatially-inhomogeneous relaxation mechanism intrinsic to the high-field ordered phase.

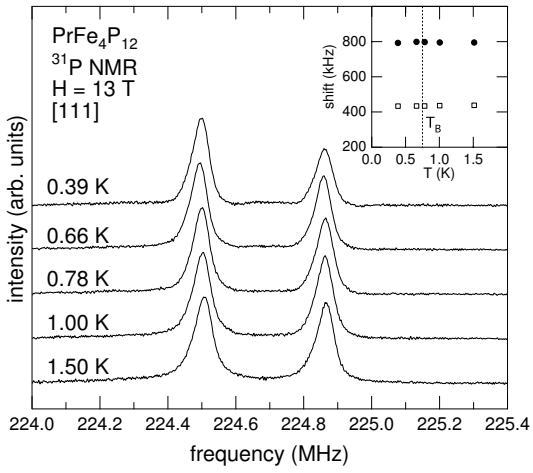

Figure 1: Temperature variation of the ^{31}P NMR spectrum at the field of 13 T along [111]. Inset: Temperature dependence of the ^{31}P frequency shifts. The dotted line in the inset shows the transition temperature $T_B \approx 0.70$ K at 13 T.

[1] D. Aoki *et al.*, J. Phy. Chem. Solids **63**, (2002) 1201.
[2] T. Tayama *et al.*, J. Phy. Soc. Jpn. **73**, (2004) 3258.
[3] J. Kikuchi *et al.*, Physica B **359-361**, (2005) 877.

PB06

Notes on the Hyperfine Interaction in Rare-earth Skutterudites

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The splitting of NMR frequency due to the AFQ ordering (SNMRF) is analyzed based on the invariant form[1] of the hyperfine interaction. The SNMRF for the P nucleus on the sites $\boldsymbol{\rho}_1 = (0, u, v)$ and $\boldsymbol{\rho}_2 = (0, u, \bar{v})$ in $PrFe_4P_{12}$ is given as ,

$$\begin{aligned}
\Delta H_{\rm hf}(\boldsymbol{\rho}_i) &= \Big[C_{1,2}(-\frac{1}{2}\bar{O}_{u,\boldsymbol{Q}} + \frac{\sqrt{3}}{2}\bar{O}_{v,\boldsymbol{Q}}) + C_{1,3}(\frac{\sqrt{3}}{2}\bar{O}_{u,\boldsymbol{Q}} + \frac{1}{2}\bar{O}_{v,\boldsymbol{Q}})\Big]\hat{H}_x^2 \\
&+ \Big[C_{2,1}(-\frac{1}{2}\bar{O}_{u,\boldsymbol{Q}} - \frac{\sqrt{3}}{2}\bar{O}_{v,\boldsymbol{Q}}) + C_{2,3}(-\frac{\sqrt{3}}{2}\bar{O}_{u,\boldsymbol{Q}} + \frac{1}{2}\bar{O}_{v,\boldsymbol{Q}})\Big]\hat{H}_y^2 \\
&+ \Big[C_{3,2}(\bar{O}_{u,\boldsymbol{Q}} - \frac{\sqrt{3}}{2}\bar{O}_{v,\boldsymbol{Q}}) + C_{3,4}(-\bar{O}_{v,\boldsymbol{Q}})\Big]\hat{H}_z^2 \\
&\pm \Big[C_{2,2}\bar{O}_{u,\boldsymbol{Q}} + C_{2,4}(-\bar{O}_{v,\boldsymbol{Q}}) \\
&\quad + C_{3,1}(-\frac{1}{2}\bar{O}_{u,\boldsymbol{Q}} - \frac{\sqrt{3}}{2}\bar{O}_{v,\boldsymbol{Q}}) + C_{3,3}(-\frac{\sqrt{3}}{2}\bar{O}_{u,\boldsymbol{Q}} + \frac{1}{2}\bar{O}_{v,\boldsymbol{Q}})\Big]\hat{H}_y\hat{H}_z \\
&+ \Big[C_{4,1}\frac{1}{2}(-\bar{O}^3_{u,\boldsymbol{Q}} + 3\bar{O}_{u,\boldsymbol{Q}}\bar{O}^2_{v,\boldsymbol{Q}}) + C_{4,2}\frac{1}{2}(-\bar{O}^3_{v,\boldsymbol{Q}} + 3\bar{O}_{v,\boldsymbol{Q}}\bar{O}^2_{u,\boldsymbol{Q}})\Big] \\
&+ [C^{(g)}_{1,2}\hat{H}_x^2 + C^{(g)}_{2,1}\hat{H}_y^2 + C^{(g)}_{3,2}\hat{H}_z^2 \pm (C^{(g)}_{2,2} + C^{(g)}_{3,1})\hat{H}_y\hat{H}_z]\Delta^{(0)}_{\boldsymbol{Q}}\,. \qquad (1)
\end{aligned}$$

Here, $\bar{O}_{u,\boldsymbol{Q}}$ and $\bar{O}_{v,\boldsymbol{Q}}$ are the AFQ order parameter of the Γ_3 type, and $\Delta_{\boldsymbol{Q}}$ is a monopole type order parameter, such as the local cubic distortion, $\hat{H}_\nu$ is the direction cosine of the magnetic field. The constants $C_{i,j}$ and $C^{(g)}_{i,j}$ are proportional to the strength of the magnetic field. The minus of $\pm$ should be used for the site $\boldsymbol{\rho}_2$. The SNMRF for other sites $\boldsymbol{\rho}_3 = (v, 0, u)$, $\boldsymbol{\rho}_4 = (v, 0, \bar{u})$, $\boldsymbol{\rho}_5 = (u, v, 0)$ and $\boldsymbol{\rho}_6 = (\bar{u}, v, 0)$ is obtained by doing the rotational operation. The SNMRF on the site $-\boldsymbol{\rho}_i$ is equal to that on the site $\boldsymbol{\rho}_i$. The figure shows the fitted result to recent SNMRF in $PrFe_4P_{12}$ studied by J. Kikuch et. al.[2]. The magnetic field dependence of AFQ is assumed as given in the lower panel which is consistent with the ND data[3]. The term $C_{4,1}$ plays important roles.

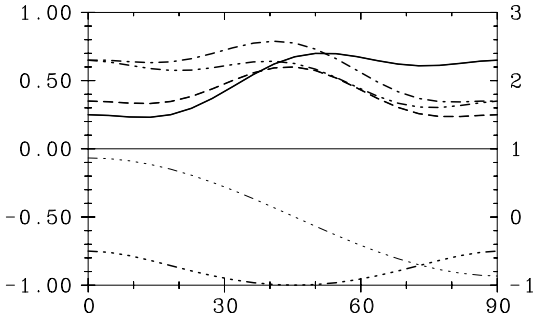

Figure 1: SNMRF when the field is rotated in the (001) plane (The horizontal axis is the angel from the x-axis). The solid line is SNMRF on site (1) and (2), the dashed line for sites (3) and (4), the dot-dashed line is for (5) and the two-dashed line for (6). The bold(thin) eight-dots-dashed line is AFQ moment of $u(v)$-type. The unit of SNMRF (left axis) is arbitrary, and right axis gives AFQ moment.

[1] O. Sakai, et al., J. Phy. Soc. Jpn. **74**, (2005) 457.
[2] J. Kikuchi, et al., (preprint and private communication) (2005)
[3] L. Hao, et al., Acta Phys. Polonica B **34** (2003) 1113.

PB07

Electrical Transport Properties in the High-Field Ordered State of $PrFe_4P_{12}$

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abstract

One of the unique properties of Pr-based filled skutterudite $PrFe_4P_{12}$ is the anomalous field-induced heavy-fermion (HF) behavior. Our focused issue is the high-field HF state especially for H applied parallel to the [111] axis, around which quite anisotropic features including the new high-field ordered state recently revealed by the thermodynamic properties [1] have been reported. In this work, we report the high-field ordered state (HOS) more closely by means of the electrical resistivity ρ and Hall resistivity ρ_H in applied H around the [111] direction. Figure 1(a) shows the temperature dependence of ρ at 14.0 and 17.9 T. A clear anomaly is observed around 0.7 K, corresponding to the phase transition to HOS. The transition temperature slightly increases with increasing H, indicating no tendency to decrease with H over the range investigated. A decrease of $\rho(T)$ below the transition temperature could be due to a reduced magnetic scattering, in contrast with the huge increase of $\rho(T)$ in the AFQ ordering [2], reflecting the Fermi surface reconstruction caused by the nesting effect [3]. Based on the anomalies found in $\rho(H)$ as well as $\rho(T)$, we determined a H-T phase diagram around the [111] direction, as shown in Fig. 1(b). New phase boundaries derived from the $\rho(H)$ measurements are found within HOS above 16 T, which are considered to be of first order.

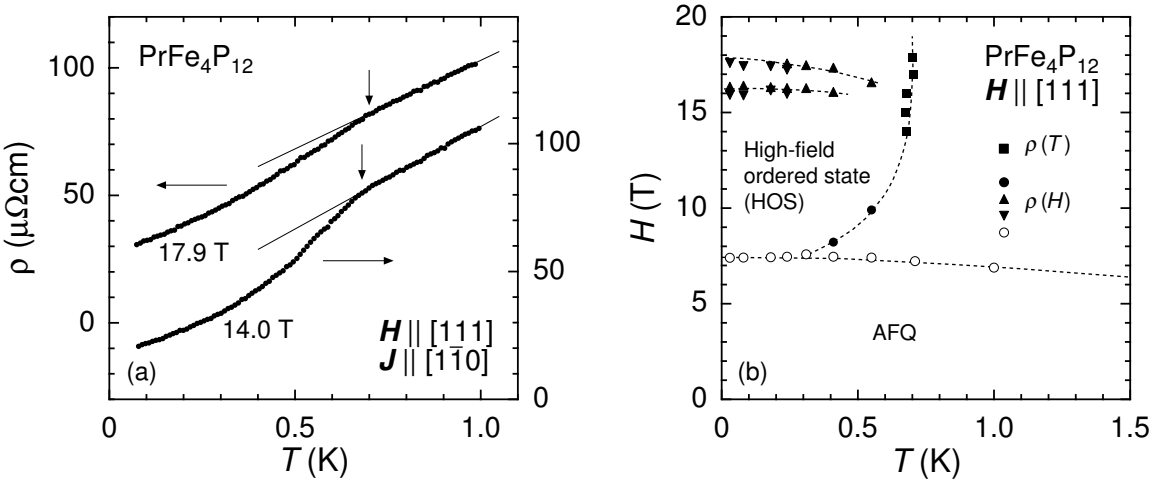

Figure 1: (a) Temperature dependence of ρ at 14.0 and 17.9 T. (b) H-T phase diagram for $H \parallel [111]$.

[1] T. Tayama *et al.*, J. Phys. Soc. Jpn. **73**, (2004) 3258.
[2] H. Sato *et al.*, Phys. Rev. B **62**, (2000) 15125.
[3] H. Harima *et al.*, Physica B **312-313**, (2002) 843.

PB08

^{31}P NMR studies of $PrFe_4P_{12}$ in a new high field phase

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Recently, a new high field phase was found under magnetic fields applied along [111] in $PrFe_4P_{12}$.[1] We have measured ^{31}P NMR in PrF_4P_{12} from 4.3K to 390mK in order to clarify the origin of the new high field phase. At the magnetic field of 13T, neither broadening nor splitting of the ^{31}P NMR spectrum was observed below the transition temperature $T_B \approx 700$mK, indicating no change of local magnetic field. However, in the measurement of the relaxation rate $1/T_1$ we found multi-componet behavior or inhomogeneous distribution of $1/T_1$ below T_B.

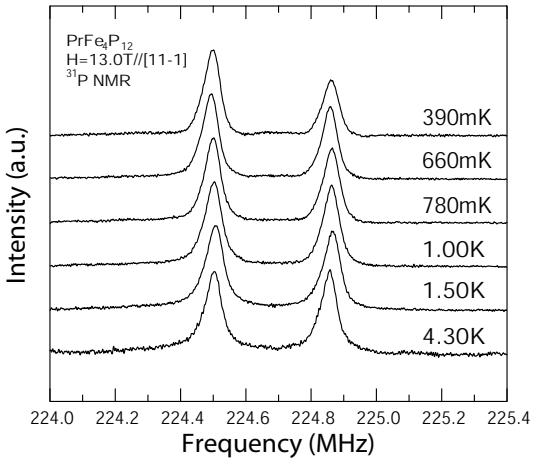

Figure 1: Temperature dependence of ^{31}P NMR spectrum under H=13T||[11-1]

[1] T. Tayama et al, J. Phy. Soc. Jpn. **73**, (2004) 3258.

PB09

Magnetic excitations in the heavy electron state of $Pr_{1-x}La_xFe_4P_{12}$(x=0 and 0.15)

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$PrFe_4P_{12}$ exhibits Kondo behavior with heavy electron mass of $81m_0$[1]. The heavy electron state was suppressed by a formation of antiferro-quadrupolar(AFQ) ordering below T_A=6.5K[2]. The ordered state is quite sensitive to the La-substitution(no AFQ order by only 15% of La-substitution)[3]. The La-substitution systems conserve the large value of Sommerfeld coefficient down to the lowest temperature. Recently, Otsuki *et al.* theoretically proposed that, under condition of small crystal field splitting between a singlet and a triplet of Pr-ion $4f^2$ state, the antiferromagnetic exchange interaction between $4f$ and conduction electrons can exist to be responsible for Kondo effect[4]. They also calculated generalized magnetic susceptibility $\chi(\omega)$ based on this model. We report quasielastic magnetic excitation spectra in the heavy electron state of $Pr_{1-x}La_xFe_4P_{12}$ (x=0 and 0.15) and compare them with the calculated $Im\chi(\omega)$. The data of x=0 was already shown in ref.5.

Circles of fig.1 shows the magnetic response of $Pr_{0.85}La_{0.15}Fe_4P_{12}$ at T=1.5K evaluated from the measured incoherent scattering intensity of vanadium and the subtraction of the phonon intensity from $LaFe_4P_{12}$. The spectrum was reproduced by a Lorentzian relaxation function with $S(\omega) = cf^2 \frac{\omega}{1-exp(-\omega/k_BT)} \frac{1}{\pi} \frac{\Gamma}{\omega^2+\Gamma^2}$ as shown by a line. This fact indicates that the hybridization between $4f$ and conduction electrons remains to the low temperature, which is consistent with the heavy electron state. Figure 2 shows experimentally determined imaginary part of magnetic susceptibility $\chi(\omega)$. These results are almost close to the calculated ones based on the singlet-triplet model.

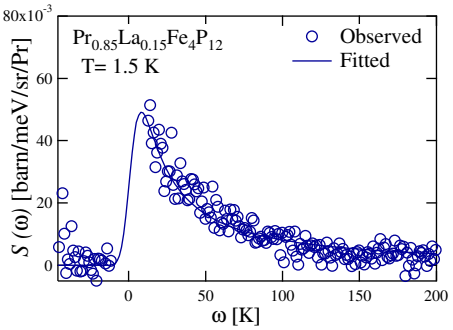

Figure 1: Magnetic excitation spectrum of $Pr_{0.85}La_{0.15}Fe_4P_{12}$ at T=1.5K.

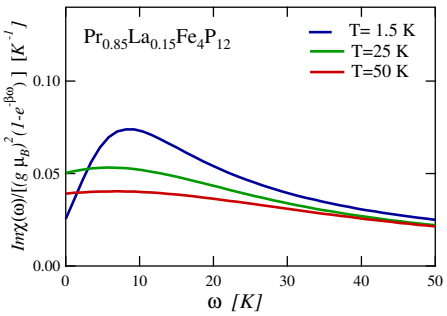

Figure 2: Imaginary part of magnetic susceptibility $\chi(\omega)$ at various temperatures.

[1] H. Sugawara *et al.*: Phys. Rev. B **66** (2002) 134411. [2] L. Hao *et al.*: Acta Phys. Polonica B **34** (2003) 1113. [3] Y. Aoki *et al.*: J. Phys. Chem. Solids **63** (2002) 1201. [4] J. Otsuki *et al.*: J. Phys. Soc. Jpn **74** (2005) 200. [5] K. Iwasa *et al.*: Acta Phys Polonica B **34** (2003) 1117.

PB10

Multipolar orderings in skutterudites

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Rare earth filled skutterudites are in the focus of interest from both theoretical and experimental sides nowadays because of their complex and intriguing behavior. Varied electronic orders of multipolar moments are realized in these systems, which are far from being fully explored yet. Quadrupolar moments seem to be the good candidate to describe the ordered phase in both $PrOs_4Sb_{12}$ and $PrFe_4P_{12}$, while the importance of octupolar moments has turn up recently in the case of $SmRu_4P_{12}$. Even higher order multipoles like hexadecapoles can be responsible for certain orderings in skutterudites, because they are possible order parameters carried by the singlet-triplet level scheme of $4f^2$ electrons, for example, contrary to the case of the quartet ground state in CeB_6.

Neutron and X-ray scattering measurements [1,2] suggest that the low-field phase in $PrFe_4P_{12}$ is the order of Γ_3 quadrupoles with ordering vector $\mathbf{q} = (1, 0, 0)$. We examine the properties of the Γ_3 quadrupoles within the singlet-triplet low-lying level scheme, which seems to be relevant to this system. We found that the antiferro-type ordered phase of these quadrupolar moments within the triplet state is quite different from the case of classical $s = 1/2$ spins or AFQ pseudo-spins within the Γ_3 doublet state. Namely, a macroscopic degeneracy is found in the ground state with respect to different quadrupolar patterns similarly to the case of the three-state antiferromagnetic Potts model. We discuss different mechanisms in order to resolve the quadrupolar degeneracy such as coupling to the lattice, further multipolar interactions or possible fluctuation effects beyond the mean-field theory.

Furthermore, we study whether the metal-insulator phase transition at $T_{\rm MI} \approx 65$K and the strange temperature-evolution of the crystalline electric field levels below $T_{\rm MI}$ in $PrRu_4P_{12}$ [3] can be characterized by the ordering of fourth- and/or six-order scalar operators.

[1] L. Hao et al., Acta Physica Polonica B **34**, (2003) 1113.
[2] K. Iwasa et al., Physica B **312-313**, (2002) 834.
[3] K. Iwasa et al., Phys. Rev. B **72**, (2005) 024414.

PB11

On the origin of metal-insulator transition in $PrRu_4P_{12}$

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Among filled skutterudite compounds, $PrRu_4P_{12}$ shows metal-insulator transition at $T_{\rm MI}$=60K [1]. From recent experiments, it has been clarified that the crystal structure of $PrRu_4P_{12}$ transforms from a body center cubic lattice for $T > T_{\rm MI}$ to a simple cubic lattice consisting of two different Pr-sites within new unit cell for $T < T_{\rm MI}$ [2,3,4]. Furthermore, it has been observed that accompanied by the structural transformation, level schemes of Pr-ions change considerably [5]. Therefore, these experimental results cause much discussion.

We study the metal-insulator transition of $PrRu_4P_{12}$ from theoretical point of view, based on j-j coupling scheme. For the metal-insulator transition, we propose antiferro-hexadecapole ordering without breaking local symmetry. Some calculated results are given in figure. This scenario provide reasonable explanation for various anomalies in $PrRu_4P_{12}$.

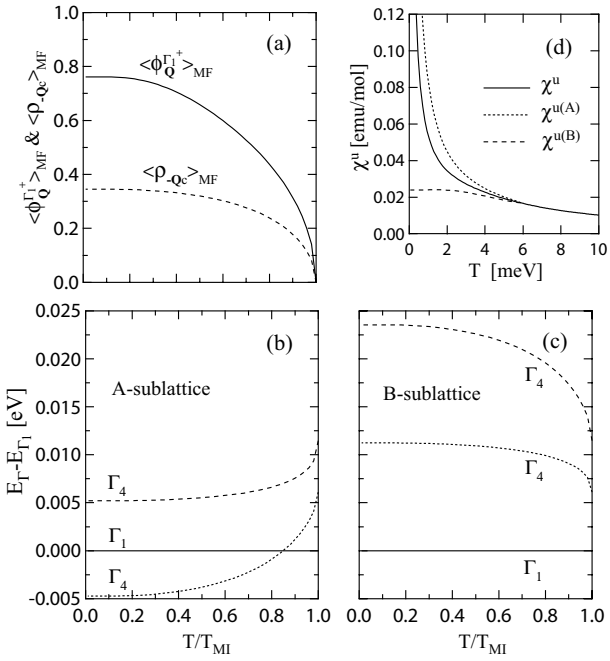

Figure 1: (a) Temperature dependences of order parameters $\langle\phi^{\Gamma_1^+}_{\mathbf{Q}}\rangle_{\rm MF}$ and $\langle\rho_{-\mathbf{Q}c}\rangle_{\rm MF}$. (b) Temperature dependence of level scheme of lowest three f^2-states in A-sublattice. (c) Temperature dependence of level scheme in B-sublattice. (d) Temperature dependence of uniform susceptibilities $\chi^{\rm u(A)}$ in A-sublattice, $\chi^{\rm u(B)}$ in B-sublattice, and $\chi^{\rm u}$ of average between A- and B-sublattices.

[1] C. Sekine, et al., Phys. Rev. Lett. **79**, (1997) 3218.
[2] C. H. Lee, et al., Phys. Rev. **B70**, (2004) 153105.
[3] D. Cao, et al., Phys. Rev. Lett. **94**, (2005) 036403.
[4] K. Iwasa, et al., J. Phys. Soc. Jpn. **74**, (2005) 1930.
[5] K. Iwasa, et al., Phys. Rev. **B72**, (2005) 024414.

PB12

Structural Analysis of $PrRu_4P_{12}$ in the Metallic Phase

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We have previously shown that $PrRu_4P_{12}$ becomes metallic and superconducting above 12 GPa [1]. At ambient pressure, it undergoes a metal-insulator transition related to a structural transition at 62 K [2-4]. Above 11 GPa, the resistivity ρ shows metallic behavior, while an anomaly in ρ at around 60 K was still observed. Two characteristic anomalies in ρ at the temperatures T_{A1} and T_{A2} became clear with increasing pressure. These facts suggest that Fermi surface nesting [5] may still exist at higher pressure, and it plays an important role in metallization.

To clarify the changes of structure at around 11 GPa, we performed powder X-ray diffraction measurements of $PrRu_4P_{12}$ under high-pressure and low-temperature. We can reproduce the observed data satisfactorily with the $Pm\bar{3}$ model, the low-temperature structure at ambient pressure [3, 4]. This result suggests the structural transition still occurs in the metallic phase and supports the gap-like behavior at around 60 K due to Fermi surface nesting at high pressure. Crystal structures in the non-metallic phase and metallic phase are shown in Fig.1. Anomalous enhancements of the P atomic displacements are observed in the high-pressure metallic phase. The displacements of P atoms are larger by one order of magnitude than in the insulating phase. These changes may play a key role in the metallization at higher pressure.

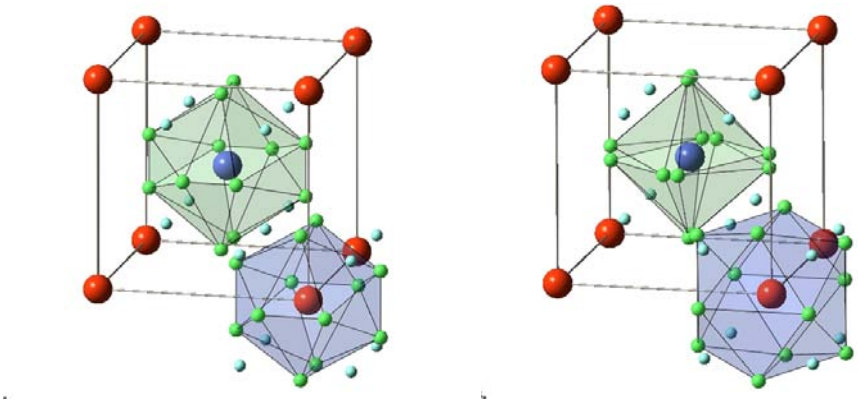

Figure 1: Schematic drawing of the crystal structure of $PrRu_4P_{12}$ in the non-metallic phase at 2.3 GPa and 10 K (left) and metallic phase at 17.0 GPa and 10 K(right). Atomic displacements, compared to the structure at 70 K, are exaggerateed by a factor of 20.

[1] A. Miyake *et al.*, J. Phys. Soc. Jpn **73** (2004) 2370.
[2] C. Sekine *et al.*, Phys. Rev. Lett. **79** (1997) 3218.
[3] C. H. Lee *et al.*, Phys. Rev. B **70** (2004) 153105.
[4] L. Hao *et al.*, J. Mag. Mag. Mater. **272-276** (2004) e271.
[5] H. Harima *et al.*, Acta Phys. Pol. B **34** (2003) 1189.

PB13

Effect of Rh substitution on metal-insulator transition compound $PrRu_4P_{12}$

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$PrRu_4P_{12}$ exhibits a metal-insulator (M-I) transition at 63K [1]. The electron and X-ray diffraction studies revealed a superlattice structure below the M-I transition temperature [2]. Recent inelastic neutron scattering experiment demonstrated the existence of two kinds of crystal-field schemes of Pr ions due to the low temperature superlattice formation and strong hybridization between 4f and conduction electrons [3]. The band calculation study suggested that the M-I transition is caused by the perfect 3 dimensional nesting [4]. However, the mechanism of the M-I transition is still an enigma. To investigate the origin of the M-I transition, the effect of Rh substitution on $PrRu_4P_{12}$ is studied by electrical resistivity, magnetic susceptibility and specific heat measurements. Single-phase polycrystalline $Pr(Ru_{1-x}Rh_x)_4P_{12}$ was prepared at high temperatures and high pressures.

Several percent Rh substitution completely suppressed the M-I transition. The transition could be due to nesting because it is extremely sensitive to a small amount of Rh substitution. Furthermore, heavy fermion state (C/T = 250 mJ/molK^2 at 2K) emerged in $Pr(Ru_{1-x}Rh_x)_4P_{12}$ which does not show the M-I transition (Fig. 1). The existence of strong hybridization between 4f and conduction electron was suggested for $PrRu_4P_{12}$.

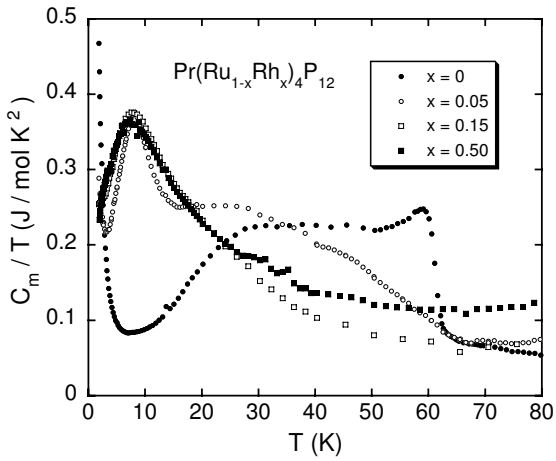

Figure 1: Specific heat C of $Pr(Ru_{1-x}Rh_x)_4P_{12}$ (x = 0, 0.05, 0.15, 0.5).

[1] C. Sekine et al, Phys. Rev. Lett. **79** (1997) 3218.
[2] C. H. Lee et al, Phys. Rev. B **70** (2004) 1531051.
[3] K. Iwasa et al, Physica B **359-361** (2005) 833.
[4] H. Harima et al, Acta Physica Plonica B **34** (2003) 1189.

PB14

Comparison of the transport properties in $R\mathrm{Ru_4P_{12}}$ (R=Pr, Sm)

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The filled skutterudite compounds $R\mathrm{Ru_4P_{12}}$ (R=Pr, Sm) are reported to be exhibit metal-insulator (M-I) transition at $T_{\mathrm{MI}} = 62$ and 16 K, respectively [1, 2]. The magnetic susceptibility in $\mathrm{PrRu_4P_{12}}$ shows no anomaly around the T_{MI} [1], and the electron diffraction experiments elucidated the structural transformation [3]. In contrast, in $\mathrm{SmRu_4P_{12}}$, magnetic susceptibility shows clear anomaly at T_{MI} [2], and no structural transformation has been reported. In order to understand the origin of these M-I transitions in $R\mathrm{Ru_4P_{12}}$ (R=Pr, Sm), we have investigated the transport properties. In this report, we present a comparison of transport properties between $\mathrm{PrRu_4P_{12}}$ and $\mathrm{SmRu_4P_{12}}$, including measurements of the temperature T dependences of electrical resistivity ρ, Hall coefficient R_{H}, and thermoelectric power S.

Figure 1 shows the T dependences of ρ in $R\mathrm{Ru_4P_{12}}$ (R=Pr, Sm). In $\mathrm{PrRu_4P_{12}}$, on cooling, ρ decreases with proportional to T and suddenly increases at T_{MI}. ρ shows a hump around 40 K, and the level crossing of the crystal electric field also occurs around 40 K [4]. In $\mathrm{SmRu_4P_{12}}$, ρ shows resistivity minimum around 50 K and shows discontinuous behavior at T_{MI}. Interestingly, $\mathrm{SmRu_4P_{12}}$ also shows hump structure in ρ below T_{MI}. In this presentation, the origin of the hump structure will be discussed.

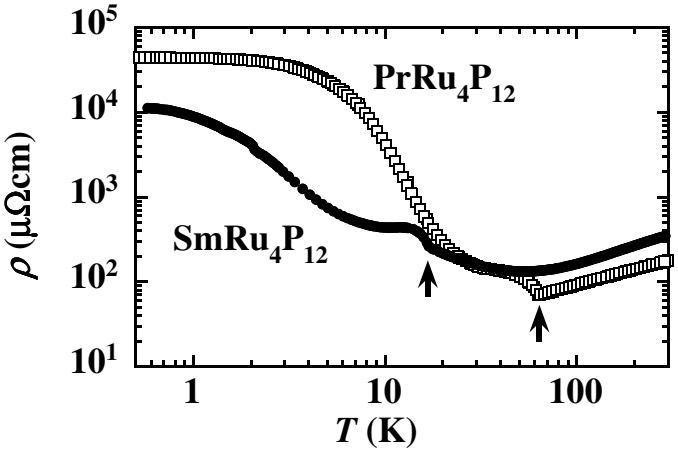

Figure 1: Electrical resistivity ρ versus temperature T for $R\mathrm{Ru_4P_{12}}$ (R=Pr, Sm). Arrows indicate $\mathrm{T_{MI}}$.

[1] C. Sekine et al., Phys. Rev. Lett. **79**, (1997) 3218.
[2] C. Sekine et al., Science and Technology of High Pressure, Universities Press, Hyderabad, India 2000, p.826.
[3] C.H. Lee et al., J. Phys.: Condens. Matter **13** (2001) L45.
[4] K. Iwasa et al., Phys. Rev. B **72**, (2005) 024414.

PB15

Elastic Properties of Filled Skutterudites of $GdRu_4P_{12}$ and $TbRu_4P_{12}$ with Heavy Lanthanids

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K.Matsuhira [1] , S.Takagi [1],C.Sekine [2] ,I.Sirotani [2]
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We have performed ultrasonic measurement on the polycrystalline filled skutterudites compounds $GdRu_4P_{12}$ and $TbRu_4P_{12}$ with heavy Lanthanids. $GdRu_4P_{12}$ and $TbRu_4P_{12}$ show a ferromagnetic transition at around 22K and 20K, respectively[1]. Furthermore another transition followed by the ferromagnetic transition appears at around 10K in $TbRu_4P_{12}$. To obtain an insight of their elastic properties and their 4f-ground states the elastic constants were measured around the phase transition. Figures 1(a) and (b) show a temperature dependence of the longitudinal elastic constant (C_L) for $TbRu_4P_{12}$ and its detailed behavior around the transition, respectively. C_L increases monotonically with decreasing a temperature. A distinct anomaly was observed at 20 K probably due to the ferromagnetic transition. A characteristic softening due to a 4f ground state of Tb ion seems not to be observed. We will present the elastic constants of $GdRu_4P_{12}$ and discuss the elastic properties around the ferromagnetic transition, comparing those of a ferromagnetic material $NdFe_4P_{12}$[2] and week ferromagnetic materials $SmFe_4P_{12}$[3] and $SmOs_4Sb_{12}$[4].

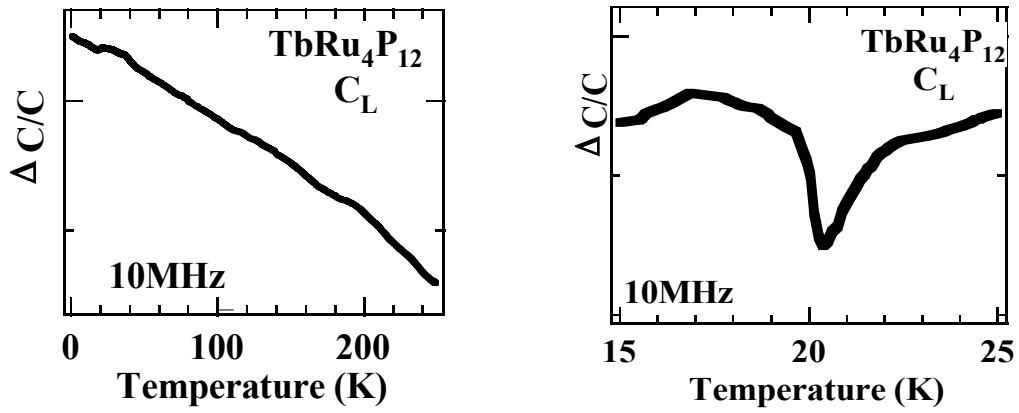

Figure.1 (a) Temperature dependence of C_L for $TbRu_4P_{12}$ and (b) its detailed behavior around 20K.

Reference

[1] C. Sekine *et al*., Phys. Rev. B **62** (2000) 11581-11584.
[2] Y.Nakanishi *et al*., Phys. Rev. B **69** (2004) 064409.
[3]S.Sanada *et al*, J. Phys. Soc. Jpn. **74** (2005) 246-249.
[4]N.Takeda and M.Ishikawa, J. Phys.: Condens. Matter **15** (2003) L229.

PB16

Single crystal growth of the filled skutterudite compounds under high pressures

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The filled skutterudite compounds RT_4X_{12}(R = rare earth, T = Fe, Ru and Os, X = P, As and Sb) have been reported to exbihit various unique properties. The samples have been synthesized by two different methods; the flux method and the high-pressure synthesis. To obtain intrinsic features of them, it is essentially important to make measurements on single crystal samples. Until now, high quality single crystals have been grown only by the flux method under ambient pressure. However, successfully grown single crystals have been limited to selected combinations of R, T, and X components by the method, and many combinations such as arsenic compounds, ROs_4P_{12} and RFe_4Sb_{12} have not yet been grown. In contrast, the high-pressure synthesis has been successfully utilized to grow filled skutterudites even for the filled skutterudites containing heavy rare earth elements by Prof. Shirotani and co-workers[1][2],although they are polycrystalline form.

Starting from the improvement of polycrystalline quality by changing growth condition, we have recently succeeded in synthesizing light rare earth filled skutterudite single crystals RFe_4Sb_{12}(R= La, Pr) by the high-pressure synthesis, although the size of single crystals is still small at this stage as shown in Fig. 1 for $PrFe_4Sb_{12}$ ($\sim$0.2mm). The crystal size keeps on increasing by changing the key factors to improve cryatal size such as the thermal gradient of sample space in furnace, the growth time, etc. The present status of the high pressure growth of the filled skutterudite single crystals will be presented.

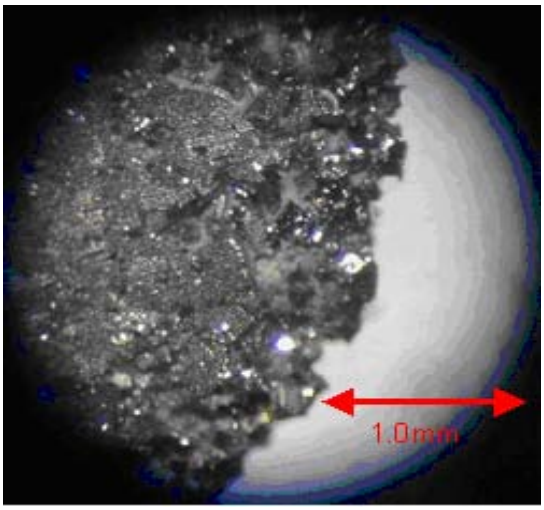

Figure 1: single crystals of $PrFe_4Sb_{12}$

[1] I.Shirotani *et al.*, J. Solid State Chem **174** (2003) 32.
[2] K.Kihou *et al.*, Materials Research Bulletin **39** (2004) 317.

PB17

Drastic effect of Pr site filling on the physical properties in $PrFe_4Sb_{12}$

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A few reports have been reported on the synthesis and physical properties of filled skutterudite compound $PrFe_4Sb_{12}$ until now [1,2,3]. In all of these reports, the samples were made under ambient pressures and Pr ion site filling factor were reported to be less than 0.87. Furthermore, those reports were inconsistent with each other on the magnetic ground state below 5 K; ferromagnetic [1], antiferromagnetic [2] and ferrimagnetic [3]. Band calculation shows that in RFe_4Sb_{12} groups (R = Rare earth), 3d electron of Fe ion have energy density of state around Fermi level uniquely [4]. Consequently, contribution to physical properties from 3d electron may be sensitive to changing Pr ion site filling factor. In order to reveal essential physical properties, it is necessary to prepare higher filling factor samples.

By utilizing the high pressure and temperature synthesis expected to increase the filling factor, we have obtained samples. From field emission electron microscope measurements, we confirmed that Pr ion sites are fully occupied. Using these samples, we have measured the electrical resistivity, the magnetic susceptibility and the specific heat. The electrical resistivity is roughly consistent with conventional reports, except the large improving of residual resistivity ratio ~ 24. Figure 1 shows the temperature dependence of M/H, exhibits a broad maximum around 10 K and the maximum temperature is roughly field independent up to 7 T. This result suggest that the broad maximum is not magnetic order. The specific heat exhibits a shotkey type peak, correspond to a broad maximum of M/H, around 10 K. The shotkey peak is ascribed to excitation of crystalline electric field Γ_1 to Γ_4, since entropy is estimated to be Rln4. $PrFe_4Sb_{12}$ is a key material to check the chang of magnetic properties for changing filling factor.

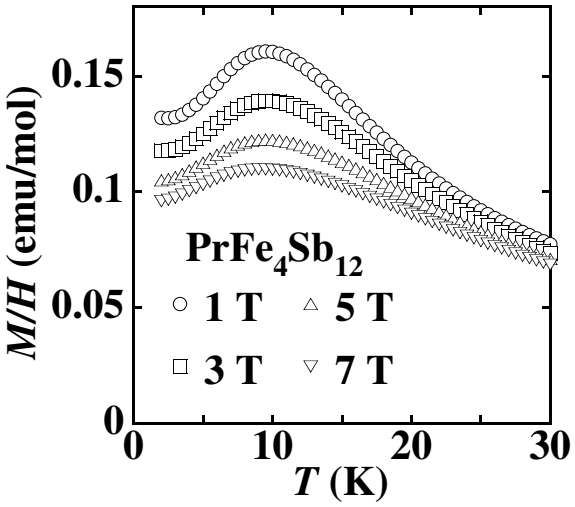

Figure 1: Temperature dependence of M/H.

[1] M.E. Danebrock *et al.*, J. Phys. Chem. Solids **57** (1996) 381.
[2] E. Bauer *et al.*, Phys. Rev. B **66** (2002) 214421.
[3] N.P. Butch *et al.*, Phys. Rev. B **71** (2005) 214417.
[4] K. Takegahara *et al.*, J. Phys. Jpn. Vol. **71** (2002) Suppl. pp. 240-242

PB18

Crystalline Electric Field State and Kondo Effect in Sm-based Filled Skutterudite Studied by Specific Heat

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Recently, Sm-based filled skutterudite compounds have been attracting much interest because of a wide variety of physical properties, such as heavy fermion (HF) behavior, metal-insulator (MI) transition, weak ferromagnetism (FM), antiferromagnetism (AFM)[1-5]. We have performed the specific heat measurement of Sm-based filled skutterudite compounds [6]. In order to reveal the origin of the wide variety of physical properties, the systematic research on Sm-based filled skutterudite compounds is necessary. We analyzed the crystalline electric field (CEF) state and the Kondo effect of $SmFe_4P_{12}$, $SmRu_4P_{12}$ and $SmOs_4P_{12}$ as compared with the NCA calculation result on single impurity Anderson model (J=5/2) [7]. The physical physical property, the CEF state and the Kondo temperature T_K of $SmFe_4P_{12}$, $SmRu_4P_{12}$ and $SmOs_4P_{12}$ are summarized in Table 1. We found that $SmFe_4P_{12}$ has the CEF ground state of Γ_5 from the analysis. On the other hand, the CEF ground state of $SmRu_4P_{12}$ and $SmOs_4P_{12}$ is Γ_{67}. We will discuss the CEF state and Kondo effect in Sm-based filled skutterudite. In addition, we will show the recent result of $SmFe_4Sb_{12}$ and $SmOs_4Sb_{12}$.

Table 1: The physical physical property, the CEF state and T_K of SmT_4P_{12}

	Property	CEF ground state	CEF level splitting	T_K
$SmFe_4P_{12}$	HF FM (T_C=1.6 K)	Γ_5 doublet	∼70 K	∼ 25 K
$SmRu_4P_{12}$	MI transition (T_{MI}=16.5 K, $T^\star$=14 K) Octupole ordering ?	Γ_{67} quartet	∼60 K	10∼15 K
$SmOs_4P_{12}$	AFM (T_N=4.5 K)	Γ_{67} quartet	∼80 K	$<$ 5 ∼10 K

[1] M.E. Danebrock *et al.*: J. Phys. Chem. Solids **57** (1996) 381.
[2] C. Sekine, *et al.*: in *Science and Technology of high Pressure*, ed M. H. Manghnant *et al.* (Univ. Press, Hyderabad, 2000) p. 826.
[3] N. Takeda and M. Ishiklawa: J. Phys.:Condens. Matter **15**, (2003) L229.
[4] R. Giri, *et al.*: Physical B **329-333** (2003) 458.
[5] S. Sanada *et al.*: J. Phys. Soc. Jpn. **74**, (2005) 246.
[6] K. Matsuhira, *et al.*: J. Phys. Soc. Jpn. **74**, (2005) 1030.
[7] A. Morishita and O. Sakai: PS34, Third Workshop on "*Skutterudite*" in Kobe Univ., Jan. 7, 2005.

PB19

Elastic properties of $SmRu_4P_{12}$ in pulsed magnetic fields

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Ultrasonic measurements in pulsed high magnetic fields have been performed for filled skutterudite compound $SmRu_4P_{12}$, which is attracting more attentions due to its mysterious origin of a metal-insulator transition at $T_{MI} = 16.5$ K and a magnetic transition at $T_N = 14$ K [1, 2]. Recently, octupolar ordering scenario was proposed to understand the successive transitions [2]. This work aims to acquire knowledge on the crystalline electric field (CEF) of $SmRu_4P_{12}$ by measuring elastic constants and ultrasonic attenuation.

Figure 1(a) shows the magnetic field dependences of the relative change $\triangle C_{11}/C_{11}$ and the attenuation β_{11} at $T = 14$ K. A distinct hardening accompanied with a sharp attenuation peak, is observed at 8 T with increasing magnetic field. This is attributed to the II-III phase transition (see H-T phase diagram in Ref.[3]). Such phase transitions were also observed at other temperatures. On the other hand, C_{44} at 4.2 K shown in the Fig.1(b) can be fitted using the reported CEF scheme that the doublet Γ_5 state located at 60 K above the ground state Γ_{67} quartet [4]. Our results indicate some differences with the reported phase diagram.

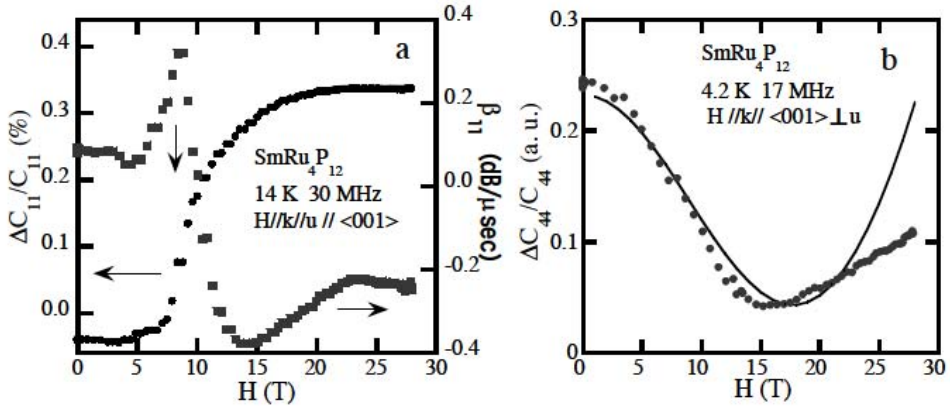

Figure 1: Magnetic field dependences of (a) the longitudinal elastic constant C_{11} and attenuation coefficient β_{11} at 14 K, and (b) the transverse elastic constant C_{44} and a fit using the CEF scheme.

[1]C. Sekine *et al.*, in *Science and Technology of High Pressure*, ed. M. H. Manghnani and M. F. Nicol (Universities Press, Hyderabad, 2000) p. 826.
[2]M. Yoshizawa *et al.*, J. Phy. Soc. Jpn. **74**, (2005) 2141.
[3]C. Sekine *et al.*, Acta Phys. Pol. B **34** (2003) 983.
[4]K. Matsuhira *et al.*, J. Phy. Soc. Jpn. **71**, (2002) Suppl., p. 237.

PB20

Spontaneous Time Reversal Symmetry Breakdown Associated with Metal-Insulator Transition in $SmRu_4P_{12}$ probed by μSR

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$SmRu_4P_{12}$ is one of the most striking compounds in filled skutterudite systems. It has been considered that an antiferro-quadrupolar (AFQ) ordering and a following antiferromagnetic ordering occur below the metal-insulator transition temperature $T_{\rm MI}$ of 16.5 K and below $T_{\rm N}$ of 15 K, respectively [1]. However the details of the order parameter especially for the phase between $T_{\rm MI}$ and $T_{\rm N}$ have not been clarified. It is recently suggested from elastic constant measurements that an octupolar ordering occurs with the spontaneous time reversal symmetry (TRS) breakdown below $T_{\rm MI}$ [2]. In order to investigate the transition, muon spin relaxation (μSR) measurements in zero and longitudinal fields (ZF and LF) have been performed at the RIKEN-RAL Muon Facility in the UK.

In ZF-μSR time spectra, a muon-spin precession was observed at low temperatures below about 5 K, which means that the ground state is a magnetically ordered state. Temperature dependence of both the initial asymmetry and the muon-spin depolarization rate exhibits an anomaly around $T_{\rm MI}$. This can be explained by a magnetic ordering below $T_{\rm MI}$, which is also supported by a result that a decoupling pattern of LF-μSR spectra indicates the occurrence of a static internal magnetic field below $T_{\rm MI}$ (Figure 1). These results concludes that the ordering below $T_{\rm MI}$ is not a non-magnetic AFQ ordering but a magnetic one with the TRS breakdown [3]. This supports a scenario that a magnetic octupolar ordering occurs below $T_{\rm MI}$ in $SmRu_4P_{12}$.

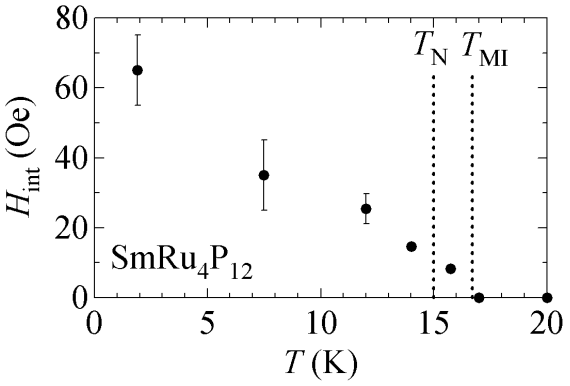

Figure 1: Temperature T dependence of the static internal field $H_{\rm int}$ estimated from the ZF- and LF-μSR.

[1] C. Sekine *et al.*, Acta Phys. Pol. B **34**, 983 (2003).
[2] M. Yoshizawa *et al.*, J. Phys. Soc. Jpn. **74**, 2141 (2005)
[3] K. Hachitani *et al.*, cond-mat/0508017.

PB21

^{31}P-NMR and μSR Studies of Filled Skutterudite System $SmRu_4P_{12}$

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In the filled skutterudite systems, $SmRu_4P_{12}$ has recently attracted much attention because of remarkable physical properties. $SmRu_4P_{12}$ was reported to exhibit a metal-insulator transition at T_{MI} of 16.5 K [1]. It have been considered that an antiferro-quadrupole (AFQ) ordering and a following antiferromagnetic ordering occur below T_{MI} and below T_N of 15K, respectively. A remarkable field-temperature (H-T) phase diagram of T_{MI} and T_N has also been reported [2]. In addition, it is recently suggested from elastic constant measurements that the ordering below T_{MI} is an octupole ordering [3]. The electronic state of this system has been studied by ^{31}P-NMR and μSR.

The line width of the ^{31}P-NMR spectrum rapidly increases below T_{MI} but not below T_{MI}. The line shape below T_N suggests a complicated magnetic structure. Temperature dependence of the spin-lattice relaxation rate $1/T_1$ below T_{MI} depends on applied magnetic fields below T_{MI}. Although an anomaly at T_N is not clear in low fields, it becomes clear above 70 kOe [4]. In 150 kOe, the successive anomaly around T_{MI} and T_N was clearly observed. The μSR experiments indicate that a static internal magnetic field appears below T_{MI}, which means that the ordering below T_{MI} is not a non-magnetic AFQ ordering but a magnetic ordering [5]. This observation supports a scenario that a magnetic octupole ordering occurs below T_{MI} in $SmRu_4P_{12}$.

[1] C. Sekine *et al.*, *Science and Technology of High Pressure*, ed. M. H. Manghnant *et al.*, 826 (Universities Press, Hyderabad, 2000).
[2] C. Sekine *et al.*, Acta Phys. Pol. B **34**, 983 (2003).
[3] M. Yoshizawa *et al.*, J. Phys. Soc. Jpn. **74**, 2141 (2005)
[4] K. Hachitani *et al.*, to be published in Physica B.
[5] K. Hachitani *et al.*, cond-mat/0508017.

PB22

NMR studies on Sm- and Yb-based filled skutterudite compounds

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We report the recent results of ^{31}P-NMR (nuclear magnetic resonance) experiments on Sm-, and Yb-based filled skutterudite compounds. $SmRu_4P_{12}$ shows a metal-insulator transition at $T_{MI} \sim 16.5$ K and another transition at $T^* \sim 14$ K [1,2]. A ^{31}P-NMR signal in paramagnetic state splits into several resonance lines below T_{MI}, and we clearly observed two different types of signals for $T^* < T < T_{MI}$: One is a pair of signals. The interval of them is almost independent of external field and does not show any significant anomaly at T^*. We therefore conclude that these signals are dominantly associated with spontaneous internal field, and AF magnetic ordering in the low T region sets on just below T_{MI}. Another signal shows strong field dependence in its shift value and disappears below T^* as indicated by arrows in Fig. 1. The observation of the latter signal suggests that some multipole ordering coexists with the long range AF magnetic order for $T^* < T < T_{MI}$. In order to examine possible coexistence of the dipole and multipole orderings, the NMR experiment by using a single crystal sample is now in progress. We also report the result on $YbFe_4P_{12}$. There is a common point between Yb- and Sm-based compounds, *i.e.* both Yb^{3+} and Sm^{3+} electronic states have one hole in $J = 7/2$ and $5/2$ mutiplets, respectively. In this context, we will discuss similarities and differences between these two classes.

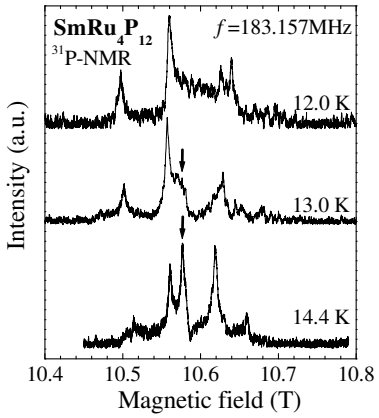

Figure 1: T variation of ^{31}P-NMR spectra below T_{MI} in $SmRu_4P_{12}$.

[1] C. Sekine *et al.*, Science and Technology of High Pressure, ed. M. H. Manghnant *et al.*, Universityes Press, Hyderabad, India, (2000) 826.
[2] K. Matsuhira *et al.*, J. Phys. Soc. Jpn. **71** Suppl. (2002) 237.

PB23

Electronic transport properties of the anomalous heavy-fermion state in $SmOs_4Sb_{12}$

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$SmOs_4Sb_{12}$ is a heavy-fermion compound, in which many interesting unconventional behaviors have been reported. The electronic specific-heat coefficientγ of $SmOs_4Sb_{12}$ is the largest among the known Sm-based compounds ($\gamma = 0.82$ J/K^{2}mol), and it doesn't show any noticeble field dependence [1]. This fact suggests that it has an unconventional origin for the heavy-fermion state formation.

We have measured the Hall coefficient and the Seebeck coefficient to investigate the anomalous state. Figure 1 shows the temperature dependence of the Seebeck coefficient $S(T)$ for $SmOs_4Sb_{12}$. $S(T)$ has a minimum at about 15K, and increases with increasing temperature. No such anomaly in $LaOs_4Sb_{12}$ suggests that it is not due to phonon drag effect but to the HF state formation. The minimum temperature of 15K can be viewed as the coherence temperature. In the similar temperature range, a change appears in the temperature dependence in T_1 obtained from Sb-NQR measurements[3]. Quite large values of $dS/dT(T = 0)$ and γ at low tempearture satisfy the empirical relation reported in HF materials[4].

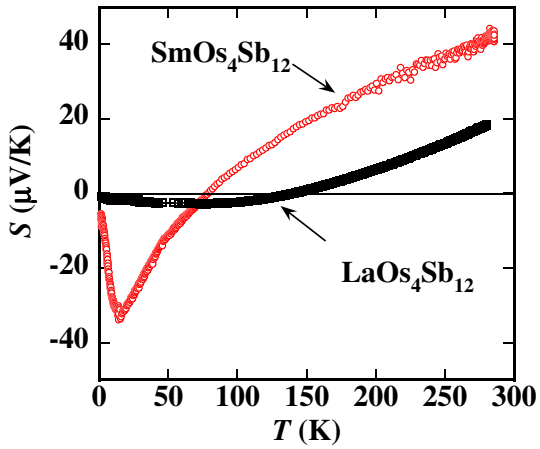

Figure 1: Temperature dependence of the Seebeck coefficient in $SmOs_4Sb_{12}$ along with $LaOs_4Sb_{12}$[2]

[1] S. Sanada *et al.* J. Phys. Soc. Jpn. **74**, (2005) 246.
[2] H. Sugawara *et al.* Phys. Rev. B **72**, (2005) 014519.
[3] H. Kotegawa *et al.* J. Phys. Soc. Jpn. **74**, (2005) 2173
[4] K. Behnia *et al.* J.Phys.:Condens. Matter **16**, (2004) 5187.

PB24

High-field magnetism of filled skutterudite compounds

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$SmOs_4Sb_{12}$ is attracted with the remarkable large γ-value of 0.82 J/K^2·mol and is regarded as the heavy fermion systems [1]. This gamma value shows no magnetic field dependence and the origin of this behavior seems to be different from the ordinary heavy fermions case. Therefore, the magnetic properties in the magnetic field is important and we measured the magnetization of the single crystal of $SmOs_4Sb_{12}$ up to 50 T below 4.2 K. Example of obtained magnetization curves are shown in the figure. The difference between 1.3 and 4.2 K at low field is due to the weak ferromagnetism with the Currie temperature of about 3 K [1,2]. There is no difference between above 15 T. The amount of the magnetization is 0.3 μ_B at 45 T which value is quite smaller than the calculated value of Sm^{+3} ion. According to Y. Aoki *et al*, the ground state is revealed by the angler dependence of the magnetization above 10 T [3].

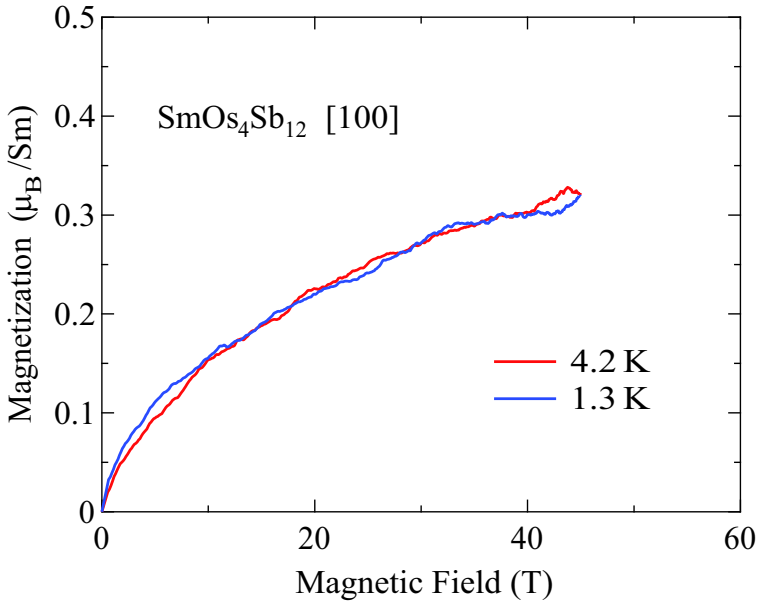

Figure 1: High-field magnetization of $SmOs_4Sb_{12}$ in the field along the [100] direction.

[1] S. Sanada *et al.*, J. Phy. Soc. Jpn. **74**, (2005) 246.
[2] W. M. Yuhasz *et al.*, Phys. Rev. B **71**, (2005) 104402.
[3] Y. Aoki *et al.*, to be published in Physica B.

PB25

Rattling Motion in the Filled Skutterudaite $SmOs_4Sb_{12}$

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One of the fascinating new experimental results in the filles skutterudaite compounds is the discovery of rattling motion due to a weakly bounded rare-earth ion in an oversized cage of pnictogen-icosahedoron. Ultrasonic measurement is one of the powerful method to explore the rattling motion. A characteristic increase appears in the temperature dependence of elastic constants originated from the Debye-type dispersion, where the incident ultrasonic wave frequency ω coincides with a relaxation time τ of the system as $\omega\tau$=1. [1] A relatively large lattice parameter in the systems with the Os_4Sb_{12} sublattice is considered for being the paradigm of rattling-motion materials. $SmOs_4Sb_{12}$ has been reported to show an unconventional heavy-fermion (HF) state, evidenced by the large specific heat coefficient γ=0.82 J/K^2 mol and the large coefficient of the quadratic temperature of the resistivity.[2] Besides, a week ferromagnetic ordering occurs below around 2.6 K.[2, 3]

Figure 1 shows the temperature dependence of the elastic constant $(C_{11}\text{-}C_{12})/2$ of $SmOs_4Sb_{12}$ with the selected frequency of 5, 17, 30 and 50 MHz. A characteristic increase was observed around 13 K. This anomaly shifts to higher temperatures and be small gradually with increasing a frequency. This behavior has a striking resemblance to that of $PrOs_4Sb_{12}$.[1] The obtained rattling parameters are presented and the detailed physical interpretation is discussed in this paper. $REOs_4Sb_{12}$ (RE: rare-earth) system is most likely to be a serious candidate to show the rattling motion and hence is appropriate to obtain the deep insight.

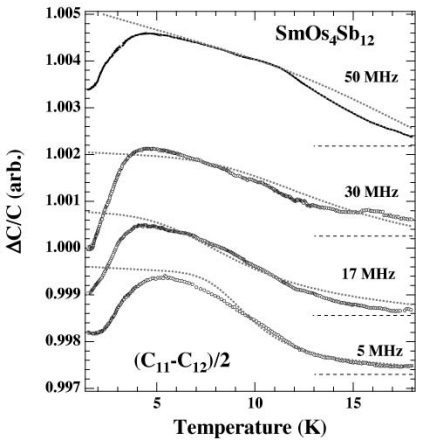

Figure 1: Temperature dependence of $(C_{11}\text{-}C_{12})/2$ of $SmOs_4Sb_{12}$ with the selected frequency of 5, 17, 30 and 50 MHz. The dotted lines are theoretical results based on the Debye-type dispersion.

[1] T. Goto *et al*, Phys. Rev. B **69**, (2004) 180511(R). [2] S. Sanada *et al*, J. Phy. Soc. Jpn. **74**, (2005) 246. [3] H. Kotegawa *et al*, J. Phy. Soc. Jpn. **74**, (2005) 2173.

PB26

Mixed valence of Sm in $SmOs_4Sb_{12}$ probed by XAS and PES

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Sm valency is considered to be quite important for the understanding of the electronic properties of Sm-based skutterudites such as $SmFe_4P_{12}$ and $SmOs_4Sb_{12}$. In order to study the Sm $4f$ electronic states, we have carried out Sm $3d$ photoabsorption spectroscopy (XAS), Sm $3d$ core-level photoemission (XPS) and valence band resonant photoemission (VB-RPES) with excitation energy of Sm $3d$ photoabsorption .

Sm $3d$ XAS of $SmFe_4P_{12}$ has a line shape which is typical of nearly trivalent Sm compounds, whereas that of $SmOs_4Sb_{12}$ has structures indicative of both divalent and trivalent states. The latter is further confirmed by bulk-sensitive Sm $3d$ XPS using ~ 8 keV incident light. Judging from the photon-energy dependence of Sm $3d$ XPS, Sm in $SmOs_4Sb_{12}$ is nearer to trivalent near the surface than in the bulk.

In the VB-RPES spectra in the Sm $3d$ photoabsorption region, structures near E_F corresponding to Sm^{2+} are found in addition to those for Sm^{3+} for both $SmFe_4P_{12}$ and $SmOs_4Sb_{12}$. Sm^{2+} state is considered to come from the surface of $SmFe_4P_{12}$ and the bulk of $SmOs_4Sb_{12}$. The finite PES intensity of Sm^{2+} state at E_F in $SmOs_4Sb_{12}$, indicates that the Sm $4f$ electronic state plays a role in the electronic states near E_F. This suggests that Sm $4f$ electrons are playing some role in the Kondo-like property of $SmOs_4Sb_{12}$. It is found that the PES intensity at E_F in $SmFe_4P_{12}$ is smaller than in $SmOs_4Sb_{12}$. This is consistent with the interpretation that mixed valency is present only near the surface in the case of $SmFe_4P_{12}$, which makes the hybridization between Sm $4f$ and pnictogen p state weaker.

PB27

Inelastic x-ray scattering study of filled-Skutterudite compounds

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abstract

Filled-Skutterudite compounds have an icosahedral cage structure consisting of twelve pnictgen atoms. In the inside of this cage, an atom such as rare-earth, actinide, alkaline, alkaline-earth and so on, is included. This characteristic crystal structure makes the presence of a localized mode expected in the inside of the cage. In fact, previous results, such as specific heat measurements, expanded x-ray absorption fine structure (EXAFS) measurements, inelastic neutron scattering, suggest the presence of a localized mode like an Einstein mode in the inside of the cage [1-3]. The presence of this mode has been believed to be crucial to understand physical properties of a series of filled-Skutterduite compounds on the aspects of not only strongly correlated electron system but also thermoelectricity, which is important for industrial application.

To understand their physical properties associated with phonon, we have applied meV inelastic x-ray scattering methods of nuclear resonant inelastic scattering (NRIS) and high resolution inelastic x-ray scattering (IXS) to samarium filled-Skutterudite compounds. The former method is useful to investigate element-specific phonon density of states [4], and the latter one is useful to investigate phonon dispersion like inelastic neutron scattering.

We have carried out ^{57}Fe and ^{149}Sm NRIS at BL09XU and IXS at BL35XU in SPring-8. We have observed sharp and periodic phonon excitations by ^{149}Sm NRIS and dispersionless modes by IXS in samarium-based filled-Skutterudite compouds. Compared the results obtained by these methods, we conclude that the samarium atoms vibrates like an Einstein mode without any assumption in samarium-based filled-Skutterudite compounds.

[1] V. Keppens et al., Nature **395**, 876 (1997).
[2] D. Cao et al., Phys. Rev B **70**, 094109 (2004).
[3] G. J. Long et al., Phys. Rev. B **71**, 140302 (2005).
[4] M. Seto et al., Phys. Rev. Lett. **74**, 3828 (1995).

PB28

Specific Heat Study on Filled Skutterudite $YbFe_4P_{12}$

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Filled skutterudite compounds LnT_4Pn_{12} (Ln = lanthanide, actinide; T = Fe, Ru, Os; Pn = P, As, Sb) have attracted attention due to their wide range of physical properties. Recently, Shirotani et al. have reported the crystal structures of new $Ln Fe_4P_{12}$ series with heavy lanthanide [1]. Among the $Ln Fe_4P_{12}$ compounds, the Ce, Eu, and Yb- based compounds show anomalies for thier lattice parameters, which is suggestive of intermediate valence states of lanthanide ion. In this study, we present an investigation of specific heat measurement for $YbFe_4P_{12}$.

Figure 1 shows the specific heat divided by temperature (C_p/T) for $YbFe_4P_{12}$. The lattice contribution $C_{lat}(LaFe_4P_{12})/T$ to the specific heat for $LaFe_4P_{12}$ is also plotted in the same figure. The electronic contribution to the C_{ele}/T for $YbFe_4P_{12}$ is represented in Fig. 2. A λ-type anomaly is found at 0.7 K, and a Schottk-type specific heat is observed around 30 K. The anomaly at 0.7 K is indicative of a long-range magnetic ordering for the Yb^{3+} moment.

In order to investigate the magnetic behavior of Yb^{3+} ions, the electronic specific heat was analized. The result reveals that the Yb^{2+} and Yb^{3+} ions coexist in the ratio of about 1 : 1. Futhermore, it is found that the ground state of Yb^{3+} is a doublet, and the energy splitting between the ground state and the first excited state is obtained to be 83 K.

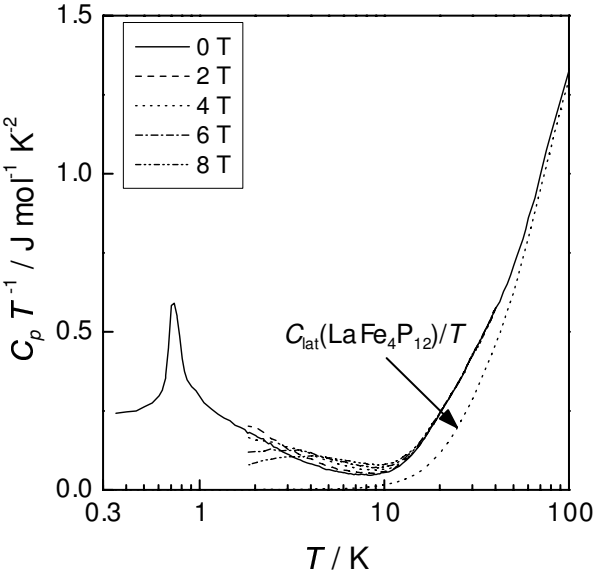

Figure 1: Specific heat divided by temperature (C_p/T) as a function of temperature for $YbFe_4P_{12}$

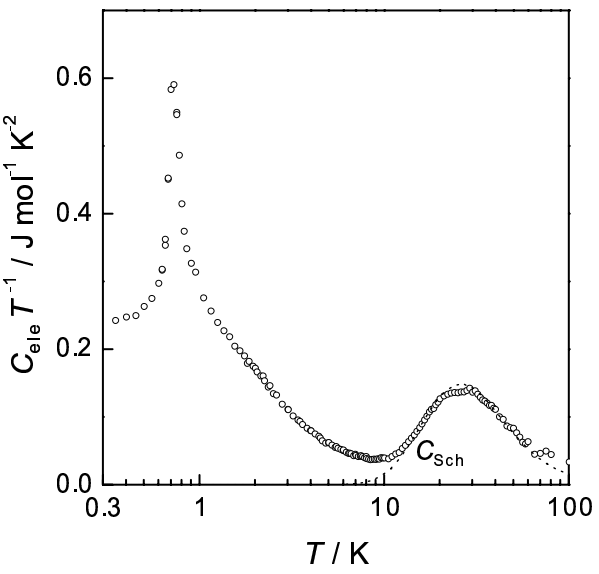

Figure 2: Temperature dependence of electronic specific heat for $YbFe_4P_{12}$.

[1] I. Shirotani et al., J. Solid State Chem. **174**, (2003) 32.

PB29

Magnetic properties of $Yb_{0.93}Fe_4Sb_{12}$

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[2] *DSM/DRECAM/SPEC, CEA Saclay, 91191 GIF/YVETTE Cedex, FRANCE*

Ytterbium is nearly divalent (valence=2.15)in $Yb_{0.93}Fe_4Sb_{12}$ and its Curie-Weiss like paramagnetic behaviour ($\mu_{eff} = 3.2\ \mu_B$.(f.u.)$^{-1}$, and $\theta_p = 50$K) arises from the itinerant electrons contributed by the [Fe_4Sb_{12}] sub-unit [1,2]. Since $NaFe_4Sb_{12}$ and KFe_4Sb_{12} ferromagnetically order below 85 K, the possibility of a transition to a weak ferromagnetic state was also initially considered in $Yb_{0.93}Fe_4Sb_{12}$. Indeed, a two orders of magnitude enhancement of the dc-susceptibilty and an irreversibility between the Zero-Field-Cooled and Field-Cooled branches were observed in weak applied field (10 Oe). However, the shape of this irreversibility was rather indicative of fine superparamagnetic particles, apparently arising from a secondary phase. To check this suggestion and definetely eliminate the possibility of a magnetically disordered state, we performed frequency dependent ac-magnetic susceptibility measurements in $Yb_{0.93}Fe_4Sb_{12}$. In a first sample, a peak centered at $T_f = 6.5$K could be observed and upon variation of the frequency, T_f was shifted with a surprisingly small variation rate $\Delta T_f/(T_f \Delta(\log\nu))$ equal to 0.016. This value is small enough to look characteristic of an intrinsic cluster glass-like behaviour in $Yb_{0.93}Fe_4Sb_{12}$. However, in samples annealed a longer time, $\Delta T_f/(T_f \Delta(\log\nu))= 0.03$ and $T_f = 4.2$K. These values are more indicative of strongly interacting fine particles and let us conclude with a good degree of confidence that $Yb_{0.93}Fe_4Sb_{12}$ is a paramagnet close to a magnetic instability.

We will also present our latest neutron and ac-susceptibility measurements on $NaFe_4Sb_{12}$.

[1] E. Alleno, D. Bérardan, C. Godart, and P. Bonville, Proceedings of SCES05 (Vienna), to appear in Physica B.

[2] W. Schnelle, A. Leithe-Jasper, M. Schmidt, H. Rosner, H. Borrmann, U. Burkhardt, J. A. Mydosh, and Y. N. Grin, Phys. Rev. B **72**, (2005) 020402.

PB30

High pressure synthesis of new filled skutterudites $R\mathrm{Rh_4As_{12}}$

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Recent improvement of systematic studies for $\mathrm{RT_4X_{12}}$ (R; rare earth, T; Fe, Ru, Os, X; P, As, Sb), the various novel physical properties such as superconductors, heavy fermions, and metal-insulator transition are realized. However, among them, only few filled skutterudites including As are studied due to the difficulty of the making sample. Using high pressure synthesizing method, new filled skutterudites $R\mathrm{Rh_4As_{12}}$ (R = La, Ce, Pr, Sm) are synthesized.

Polycrystalline $R\mathrm{Rh_4As_{12}}$ were prepared at high temperatures and high pressures using wedge-type cubic-anvil high-pressure apparatus. The compounds were prepared by the reaction of stoichiometric amounts of each metal and arsenic powders at around 950 ℃ and 2.0 GPa. The samples were characterized by powder X-ray diffraction using Cu K_α radiation and silicon as standard.

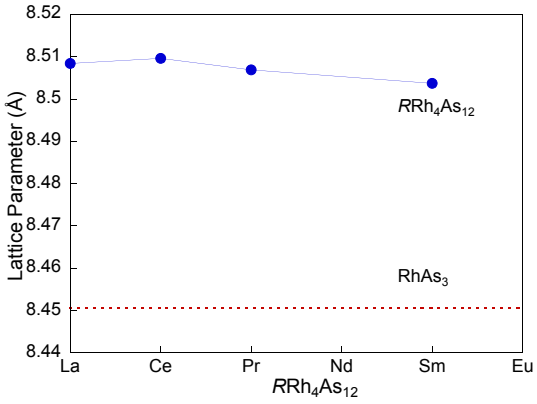

Figure 1: Lattice parameters of filled skutterudites $R\mathrm{Rh_4As_{12}}$. The dotted line is $\mathrm{RhAs_3}$.

$R\mathrm{Rh_4As_{12}}$ maintains the cubic-filled skutterudite-type structure. Fig. 1 shows the rare earth dependence of the $R\mathrm{Rh_4As_{12}}$ and unfilled skutterudite $\mathrm{RhAs_3}$. From the X-ray diffraction, the lattice parameters are determined as 8.5084 Å, 8.5096 Å, 8.5069 Å, and 8.5037 Å for $\mathrm{LaRh_4As_{12}}$, $\mathrm{CeRh_4As_{12}}$, $\mathrm{PrRh_4As_{12}}$, and $\mathrm{SmRh_4As_{12}}$. These values are systematically decreased due to the lanthanide contraction. The lattice parameters of $R\mathrm{Rh_4As_{12}}$ are $\sim$ 0.8 % larger than that of $\mathrm{RhAs_3}$ [1], which indicates the doping of rare earths to the $\mathrm{RhAs_3}$. However, rare earth is not fully doped to $\mathrm{RhAs_3}$. For the $\mathrm{LaRh_4As_{12}}$, La is $\sim$ 70 % doped to the $\mathrm{RhAs_3}$ [2].

The temperature dependence of electrical resistivity for $\mathrm{LaRh_4As_{12}}$ is decreased with decreasing temperature and saturated at low temperatures. This metallic behavior is contrast with the one of $\mathrm{RhAs_3}$, which shows the semiconducting behaviors [3].

[1] A. Kjekshus *et al.*, Acta Chem. Scand. **28** (1974) 99.
[2] K. Takeda, private communication.
[3] K. Takegahara *et al.*, Physica B **328** (2003) 74.

PB31

High-pressure and high-temperature synthesis of skutterudite compounds containing Ge in the framework.

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The filled skutterudite structure contains icosahedral cages composed of 15 group elements, in which guest atoms like rare-earth metals are situated. A great many compounds with this structure were reported, and their physical properties were extensively studied from the interest in the strange behavior of their f electrons. However, the compounds containing other group elements substituted for the 15 group elements of the host network have not been well studied. In the present work, we have tried to prepare new skutterudite compounds containing Ge in the host network using high-pressure and high-temperature reactions.

Powders of Rh (Nilaco 99.9%), Sb (Katayama Chemical 99.999%), Ge (Mitsuwa Pure Chemical 99.999%), and blocks of La (Furu-uchi Chemical 99.9%), were mixed with an atomic ratio of La : Rh : Sb : Ge = 1 : 4 : 9 : 3 in an Ar-filled glovebox. The mixture was put into an h-BN cell (5 mm in inner diameter and 5 mm in depth). The cell was covered with a tantalum foil heater, and was put in an MgO octahedron. It was placed in the eight truncated tungsten carbide cubes, and was pressed at 7 GPa and heated at 800°C for 1 hour, followed by quenching to room temperature. After the reaction the pressure was gradually released to the ambient pressure.

X-ray powder diffraction pattern of the product was measured with an X-ray diffractmeter (Bruker, D8 Advance) using CuK α radiation. The diffraction pattern of the product is shown in Figure 1. All peaks can be indexed with a cubic unit cell with a = 9.112(1) Å. From the comparison with the simulated pattern assumed the skutterudite structure, the compound was identified as a new skutterudite comopund, $LaRh_4Sb_9Ge_3$. The smaller lattice constant than those of $RhSb_3$ (9.2322 Å) [1] and $La_{0.1}Rh_8Sb_{24}$ (9.2213 Å) [2] showed that Ge were successfully substituted for a part of Sb atoms. $LaRh_4Sb_9Ge_3$ were metallic, and did not show superconductivity below to 2 K. This is the first clear example of skutterudites containing Ge partially replacing 15 group atoms in the framework.

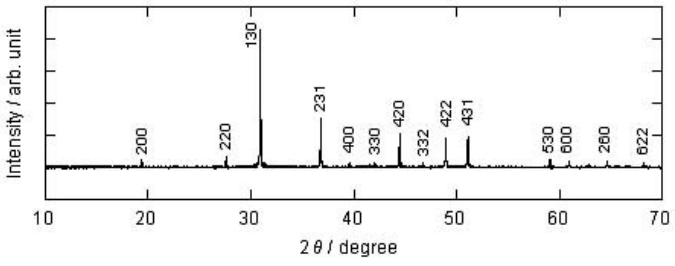

Figure 1: X-ray powder diffraction pattern of $LaRh_4Sb_9Ge_3$.

[1] A. Kjekshus, T. Rakke, Acta Chem. Scand. **A28** (1974) 99-103.
[2] L.Zeng, H.F.Franzen, J. alloys compd. **311** (2000) 224 - 225.

PB32

Clathrates and clathrate-like compounds of europium and barium

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Our recent investigations were focussed mainly on intermetallic clathrates Ba_6Ge_{25} [1,2], its Eu-substituted derivative $Ba_{6-x}Eu_xGe_{25}$ [3] and $Eu_8Ga_{16-x}Ge_{30+x}$ (α- and β-modifications) [4-6], and their derivatives. Additionally, $EuGa_{2-x}Si_{4+x}$ [7] – a clathrate-like phase – was taken into account. Several correlations of the physical properties and chemical interaction in dependence on temperature and pressure were established and characterized in detail. The temperature behaviour of cubic germanide Ba_6Ge_{25} was investigated both on polycrystalline samples and single crystals. The temperature dependence of the lattice parameter exhibits two anomalies at about 180 K and 230 K, respectively, which are caused by a structure transformation in two steps with hysteresis. Powder (T = 10 K–295 K) and single-crystal (T = 95 K–295 K) X-ray diffraction studies confirm that the symmetry of Ba_6Ge_{25} (space group $P4_132$) remains unchanged within the entire temperature range. A reconstructive behaviour of the structural transformation is observed, involving Ge–Ge bond breaking and barium cation displacements. Some Ge atoms (ca. 28% of Ge4) are so significantly displaced during cooling that the according Ge–Ge bonds break and new three-bonded (3b)Ge species (electron acceptors) are formed. Consequently, the number of charge carriers is reduced, affecting the physical properties. The reversible bond breaking involved in this process is a typical characteristic of a solid-state chemical reaction. $EuGa_{2-x}Si_{4+x}$ is prepared by high pressure-high temperature treatment and crystallizes in the structure type of $EuGa_2Ge_4$. Four-bonded gallium and silicon atoms form a 3D polyanionic network with europium cations embedded in large cavities. The electron balance can be written as $Eu^{2+}[(3b)Ga^{1-}]_{2-x}[(4b)Si^0]_{4+x}\ xe^-$. This suggests metal-like behaviour and europium atoms in the configuration $4f^7$ (Eu^{2+}). Both are confirmed by physical measurements.

[1] W. Carrillo-Cabrera *et al.* J. Solid State Chem 178, 715 (2005).
[2] I. Zerec *et al.* Phys. Rev. B 72, 045122 (2005).
[3] J. Sichelschmidt *et al.* Eur. Phys. J. B46, 201 (2005).
[4] V. Pacheco *et al.* Phys. Rev. B 71, 165205 (2005).
[5] A. Bentien *et al.* Phys. Rev. B 71, 165206 (2005).
[6] J. Sichelschmidt *et al.* Eur. Phys. J. B46, 363 (2005).
[7] A. Wosylus *et al.* XX Congress of IUCR, Book of Abstracts, C-462 (2005).

PB33

Physical Properties of New Cage-structure Compounds $R_2Rh_{15}Si_7$ and $R_3Pt_{23}Si_{11}$

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The recent discovery of heavy fermion superconductivity in $PrOs_4Sb_{12}$ has triggered intensive studies on f-electron compounds with filled-skutterudite structure.

Aiming at the search for a new cage-structure compounds, we synthesized polycrystalline sample of $R_2Rh_{15}Si_7$ and $R_3Pt_{23}Si_{11}$ by arc melting, and measured magnetic susceptibility, specific heat and electrical resistivity.

$R_2Rh_{15}Si_7$ crystallizes in the cubic structure with the space group Pm-$3m$. There are two independent R sites having O_h and D_{4h} symmetry, respectively. In $Ce_2Rh_{15}Si_7$, the magnetic susceptibility shows that the $4f$ electron is in the valence-fluctuating state. In $Pr_2Rh_{15}Si_7$, the magnetic susceptibility does not show phase transition down to 0.3 K and it still increases at the lowest temperature measured (Figure 1). The temperature dependence of the specific heat shows two broad humps at 20 K and 2 K due to the cystalline field splitting of the Pr-$4f$ state in each Pr site. In addition, a huge increase of C/T is found below 1 K, suggesting the magnetic Pr-$4f$ ground state and the onset of a phase transition (Figure 2).

$R_3Pt_{23}Si_{11}$ crystallizes in the cubic structure with the space group Pm-$3m$ and there is only one independent R-site with D_{4h} symmetry. The magnetic properties of $Pr_3Pt_{23}Si_{11}$ are very similar to those of $Pr_2Rh_{15}Si_7$, having the magnetic ground state and the increase of C/T.

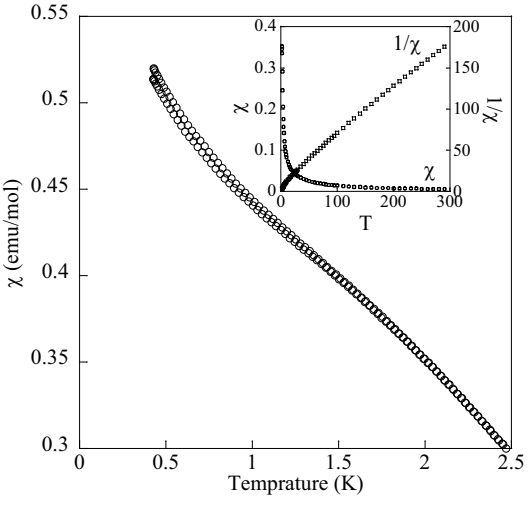

Figure 1: Temperature dependence of Magnetic susceptibility for $Pr_2Rh_{15}Si_7$

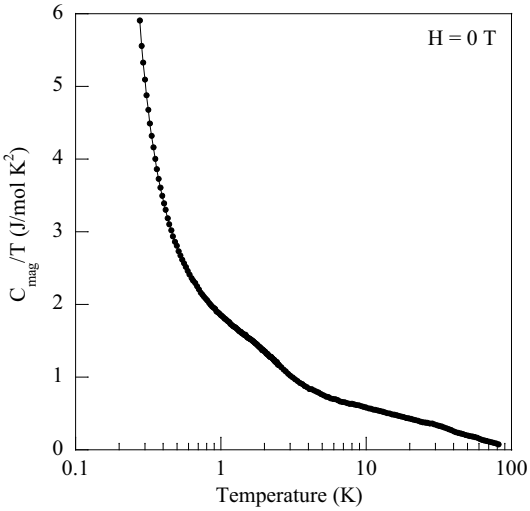

Figure 2: Temperature dependence of C_{mag}/T for $Pr_2Rh_{15}Si_7$

PB34

Single crystal growth of plutonium compounds

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The discovery of the high-temperature superconductivity in $PuCoGa_5$[1] and $PuRhGa_5$[2] accelerated research activity in trans-uranium compounds. We report recent progress in Japan of sample preparation and physical property measurements on plutonium compounds. Single crystals of plutonium compounds such as superconducting $PuRhGa_5$, antiferromagnetic $PuGa_3$ and paramagnetic $PuIn_3$ were successfully grown by the self-flux method and encapsulated for the measurements at Ar-circulated glove boxes in the Oarai reseach establishment, Japan Atomic Energy Agency. Physical properties measurements were performed at the Oarai branch, IMR, Tohoku university, which is located close to the sample preparation facility. To avoid the self-heating effect due to the strong radiation, a specially designed encapsulation was used to keep good thermal contact between the sample and the cryostat.

Using those crystals, anisotropy in the superconducting state was revealed for $PuRhGa_5$.[3] In $PuIn_3$, we succeeded in observing de Haas-van Alphen effect for the first time and revealed $5f$-itinerant electronic states. [4]

[1] J. L. Sarrao, L. A. Morales, J. D. Thompson, B. L. Scott, G. R. Stewart, F. Wastin, J. Rebizant, P. Boulet, E. Colineau and G. H. Lander, Nature (London) **420** (2002) 297.
[2] F. Wastin, P. Boulet, J. Rebizant, E. Colineau and G. H. Lander, J. Phys.: Condens. Matter **15** (2003) S2279.
[3] Y. Haga, D. Aoki, T.D. Matsuda, K. Nakajima, Y. Arai, E. Yamamoto, A. Nakamura, Y. Homma, Y. Shiokawa and Y. Ōnuki: J. Phys. Soc. Jpn. **74** (2005) 1698.
[4] Y. Haga, D. Aoki, H. Yamagami, T.D. Matsuda, K. Nakajima, Y. Arai, E. Yamamoto, A. Nakamura, Y. Homma, Y. Shiokawa and Y. Ōnuki: to be publised in J. Phys. Soc. Jpn. **74** (2005).

PB35

Single crystal growth of uranium and thorium compounds

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In order to investigate the magnetic and electronic properties of the rare earth and actinide compounds, it is essentially important to prepare the high-quality samples. Especially, to obtain a single crystal of actinide compounds, it is hard to determine an appropriate method and conditions such as temperature, starting ratio of elements, *etc.* Recently, we have been intensively developing the flux method to grow high quality single crystals of thorium and uranium compounds and succeeded to obtain those single crystals. Figure (a),(b),(c) and (d) show the single crystal of $ThIn_3$, $ThRhIn_5$, UCu_2Si_2 and UCu_2Ge_2, respectively. For $ThIn_3$, we have observed the de Haas-van Alphen signal and investigated the Fermi surface properties.[1] In the case of UCu_2Si_2, though many experimental studies were done by using polycrystal and/or low-quality single crystal samples, the magnetic properties were controversial. On this point, we have finally succeeded to clarify the magnetic properties precisely by using our single crystal.[2] We will also report our recent results of synthesis on actinide filled skutterudite.

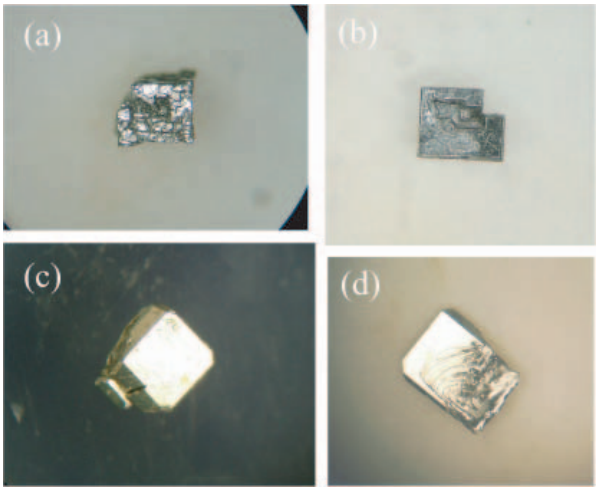

Figure 1: Single crystal of (a)$ThIn_3$, (b)$ThRhIn_5$, (c)UCu_2Si_2 and (d)UCu_2Ge_2.

[1] T.D. Matsuda, D. Author, J. Phy. Soc. Jpn. **74**, No.12 (2005).
[2] T.D. Matsuda, Y. Haga, S. Ikeda, A. Galatanu, E. Yamamoto, H. Shishido, M. Yamada, J. Yamaura, M. Hedo, Y. Uwatoko, T. Matsumoto, T. Tada, S. Noguchi, T. Sugimoto, K. Kuwahara, K. Iwasa, M. Kohgi, R. Settai and Y. Ōnuki, J. Phy. Soc. Jpn. **74**, (2005) 1552.

PB36

Non-enhancement of thermoelectric-power coefficient of $PrInAg_2$ at low temperatures

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The thermoelectric power, S, of $PrInAg_2$ has been measured at temperatures down to 0.1 K to clarify that the thermoelectric-power coefficient, S/T, is enhanced at low temperatures. The specific-heat coefficient, C/T, of this compound is increased at low temperatures with decreasing temperature [1]. Thus S/T of $PrInAg_2$ is expected to increase at low temperatures with decreasing temperature, because the S/T and C/T are well known to be strongly related in the series of the heavy-fermion compounds such as Ce-based and Yb-based Kondo compounds [2,3].

The experimental result of S of $PrInAg_2$ are shown in Fig. 1. The S of $PrInCu_2$ and $LaInCu_2$ are also shown for comparison [4]. The value of S/T of $PrInAg_2$ is -0.5×10^{-8} V/K^2 at low temperatures, the absolute value of which is less than that of $PrInCu_2$. There is no indication of the enhancement of S/T at low temperatures.

The value of C/T of $PrInAg_2$ is 6.5 $J/mol{\cdot}K^2$ at low temperatures [1]. Thus, the present experiment indicates that the relationship between S/T and C/T is broken for this compound. In the case of $PrInCu_2$, the relationship is also broken [4]. Therefore, it is concluded that the density of state and the relaxation time of conduction electrons at the Fermi energy are not modified by the Pr $4f^2$ electrons in $PrInAg_2$. Moreover, the enhancement of C/T does not mean the $PrInAg_2$ is a member of the heavy-fermion compounds.

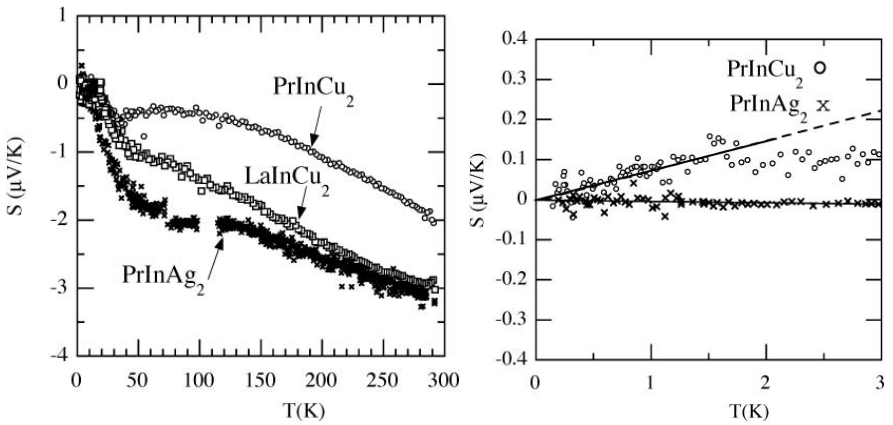

Figure 1: Thermoelectric power, S, of $PrInAg_2$ at temperatures below 300 K (left) and at low temperatures (right). The S of $PrInCu_2$ and $LaInCu_2$ are also shown for comparison.

[1] A. Yatskar, W. P. Beyermann, R. Movshovich, P. C. Canfield, Phys. Rev. Lett. **77** (1996) 3637.
[2] J. Sakurai, in *Transport and Thermal Properties of f-Electron Systems*, ed. G. Oomi *et al.* (Plenum Press, New York, 1993) p. 165.
[3] K. Behnia, D. Jaccard and D. Flouquet, J. Phys.: Condens. Matter **16** (2004) 5187.
[4] T. Ikeno, Y. Isikawa, T. Kuwai, A. Mistuda and T. Mizushima, Proceedings of SCES05 in Vienna.

PB37

Possible Evidence for Quadrupole and Octupole Fluctuations in $PrAg_2In$ with the Cubic Γ_3 Ground State ; NMR Studies

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The most striking feature of $PrAg_2In$ is that the crystalline-electric field (CEF) ground state is a non-magnetic, non-Kramers doublet Γ_3 [1,2], which has no magnetic dipole moments but has quadrupole moments O_{20} and O_{22}. In addition, with the large CEF splitting of about 70 K between the ground state and the first excited state Γ_4, and also with sharp CEF transitions, the low-T and low-frequency phenomena are not affected by magnetic dipolar degrees of freedom. Thus, $PrAg_2In$ has been considered as an ideal system in studying novel properties arising from degrees of freedom of Γ_3. Here, magnetic contributions with the CEF splitting consist of the high-frequency and low-frequency processes. Knight shift K and magnetic susceptibility χ are affected by both processes, but $1/T_1$ is affected by only low-frequency processes. Therefore, it is considered that the multipolar fluctuations might be able to be observed through the nuclear spin relaxations. We note that Γ_3 also has an octupole moment T_{xyz} in addition to quadrupole moments O_{20} and O_{22}. We have been investigating microscopic properties of $PrAg_2In$ by complimentarily utilizing ^{115}In (I=9/2) and ^{109}Ag (I=1/2) NMR on powder samples obtained by crushing single crystals grown by the Bridgman method. Our main resluts of ^{115}In and ^{109}Ag NMR are summarized as follows.

(1) The intrinsic χ evaluated from the ^{115}In Knight shift, ^{115}K shows a slight but significant increase below $\simeq$ 20 K. This behavior is not expected at all for the usual magnetism of Γ_3 ground state system, and suggests the existance of the low-frequency magnetic fluctuations even at low T. [3]

(2) The nuclear longitudinal relaxation rate of ^{115}In, $1/^{115}T_1$, shows an anomalous enhancement below $\simeq$ 50 K over the one that is expected from the low-frequency magnetic fluctuations of the excited magnetic Γ_4 and Γ_5 states, which indicates a possible contribution to $1/^{115}T_1$ from quadrupolar fluctuations of the Γ_3 ground state at low T.

(3) Contrary to a naive expectation that with I=1/2, $1/^{109}T_1$ of ^{109}Ag does not include such a "quadrupolar" contribution, $1/^{109}T_1$ shows a larger enhancement than $1/^{115}T_1$ about $\simeq$ 100 K. With similarly linear K vs χ behavior between ^{115}In and ^{109}Ag down to low T, the behavior cannnot be explained by assuming that the anomalous $1/T_1$ is dominated by the low-frequency magnetic fluctuations as revealed by the increasing intrinsic χ deduced from ^{115}K. It rather suggests, in conjunction with characteristic band structure of compound, that the Γ_3 ground-state octupolar and quadrupolar fluctuations make, respectively, significant contributions to $1/^{109}T_1$ and $1/^{115}T_1$ at low T.

[1] R. M. Galera *et al.*, J. Less-Common Metal **53** (1982) 2158.
[2] T. M. Kelley *et al.*, Phys. Rev. B **61** (2000) 1831.
[3] H. Tanida *et al.*, presented in SCES'05 vienna, to be published in Physica B.

PB38

Magnetic / Quadrupolar Properties in $PrMg_3$ with the Cubic Γ_3 Ground State

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In recent years, anomalous phenomena arising from the degrees of freedom of the quadrupole moments attract many researchers, and those are investigated energetically. In the crystalline-electric field (CEF), a ground manifold of Pr^{3+} J=4 at the cubic symmetry sites splits into a singlet Γ_1, a doublet Γ_3 and two triplets Γ_4 and Γ_5. Here, the Γ_3 is a non-magnetic, non-Kramers doublet, and it has no magnetic dipole moment, but has quadrupole moments O_{20} and O_{22}. If a cubic Pr-based compound is a system with a cubic Γ_3 ground state, there exists the possibility to show novel properties arising from the degrees of freedom of ground state Γ_3.

In this point of view, $PrMg_3$ is of great interest. From the previous inelastic-neutron scattering studies, $PrMg_3$ is characterized as a system with a cubic Γ_3 ground state and CEF level scheme Γ_3-Γ_4(56 K)-Γ_1(135 K)-Γ_5(184 K) [1,2]. Although the ground state has a degeneracy of doublet, there is no phase transition at low-T [1,3] on polycrystalline samples down to 2 K. Thus, the mechanism of releasing the degeneracy of the doublet is not clarified yet at all.

We present the resistivity ρ, magnetic susceptibility χ and specific heat C_p of $PrMg_3$ on single crystals down to 0.5 K. Samples were prepared by Bridgman method with Mo crucible. Starting materials are 4N purity, respectively. In these measurements, we can not find clear evidence corresponding to the phase transition down to the lowest T. However C_p shows dramatic increase and C_p/T reaches $\sim$4 J/mol K^2 at 0.5 K. More detailed results will be presented in the workshop.

[1] A. Andreeff *et al.*, Phys. Stat. Sol. **98** (1980) 283.
[2] R. M. Galera *et al.*, J. Magn. Magn. Mater. **23** (1981) 317.
[3] K. H. J. Buschow, J. Less-Common Metals **44** (1976) 301.

PB39

Single-Crystal ^{29}Si NMR Studies of Hidden Order in URu_2Si_2

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The still unclarified hidden order phase (HO) below T_0=17.5 K in URu_2Si_2 has been investigated on two single crystals under ambient P by ^{29}Si (I=1/2) NMR with $H_{\rm ext}$ // [100](=a), [110] and [001](=c) axes. Our major findings are as follows.

(1) To describe the tiny moment antiferromagnetism (AF) under ambient P, the model of the "Small Moment AF" (SMAF) has been proposed previously [1], which interprets the ambient P tiny moment AF in terms of spatially homogeneous AF with $\mu_{\rm SMAF}$=0.02-0.04$\mu_{\rm B}$ // [001] and $\vec{Q}_0$=[001] over the entire sample. From the observation at T=4.5 K for $H_{\rm ext}$ // c-axis of no splitting of ^{29}Si NMR lines of the order of magnitude expected from $\mu_{\rm SMAF}$, we can definitely exclude the SMAF model. Our results support the "inhomogeneous"model that the ambient P tiny moment AF occurs spatially inhomogeneously only in a very tiny part (of the order of 1% or less) of the sample, and the HO occurs in the remaining majority of the sample.
(2) In the HO state, the ^{29}Si NMR linewidth can include the additional contribution $\Delta H_{\rm HO}$ (FWHM) from internal hyperfine field arising from HO over the inhomogeneous width αM that is proportional to bulk magnetization M. Our $\Delta H_{\rm HO}$ is smaller than those in any other previous reports [2-4]. $\Delta H_{\rm HO,a}\simeq$1 Oe at T=4.6 K is about 20 times smaller than that reported on field-aligned powders by Bernal *et al.* [2]. We have $\Delta H_{\rm HO,c}\simeq$0 Oe at T=4.5 K, whereas almost isotropic $\Delta H_{\rm HO,c}$=22 Oe was reported on the same field-aligned powders [2]. We consider that the smaller $\Delta H_{\rm HO}$ and its reproducibility between two single crystals will not constitute a bad sign on our sample quality.
(3) $\Delta H_{\rm HO}$ appears below T_0 for all three $H_{\rm ext}$ directions, isotropically in the c-plane, but anisotropically between c-axis and c-plane. With I=1/2 of ^{29}Si, this indicates that the time-reversal symmetry is broken at T_0.
(4) Surprisingly, $\Delta H_{\rm HO,c}$ disappears almost completely below $\sim$14 K. This strongly suggests that the HO that sets in at T_0 does not develop monotonically with decreasing T, and may correspond well to our recent observation of the anomaly in the ^{101}Ru NQR frequency around T_Q=13.5 K, which can be interpreted as arising from symmetry-breaking electric-field-gradient components of V_{yz} and/or V_{zx} at the Ru site due to quadrupole order below this T [5]. The H0 phase is considered to hide sub-phases.
(5) $H_{\rm ext}$-dependence of $\Delta H_{\rm HO,a}$ and almost isotropic behavior of $\Delta H_{\rm HO}$ between [100] and [110] under $H_{\rm ext}$=6.7 T are also discussed in terms of possible octupole/quadrupole orders.

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Theory of de Haas-van Alphen effect in dilute Kondo systems

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Spin split effective mass has been observed in de Haas-van Alphen (dHvA) experiments of heavy fermion systems. It is remarkable that one of spin components cannot be detected in CeB_6[1]. Similar behavior has been found in $Ce_xLa_{1-x}B_6$ for $x \gtrsim 0.05$, in which effective mass of observed spin component is proportional to Ce concentration x [2]. Extremely different effective mass is one of the possible reason for the lack of one spin component in the dHvA signals. We investigate spin dependence of effective mass in dilute Kondo systems with and without orbital degeneracy.

Many body effects in dHvA oscillations are taken into account through the self-energy part of conduction electrons in the Lifshitz-Kosevich formula [3-5]. We first dicuss on constructions of the self-energy in dilute systems from that in single impurity systems. It is fundamental and crucial problem especially in Kondo systems [5, 6].

We employ single impurity SU(4) Coqblin-Schrieffer model simulating $Ce_xLa_{1-x}B_6$, and study the original Kondo model for reference. Explicit results will be shown for spin dependent effective mass and level shift computed in the numerical renormalization group (NRG).

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Interpretation of resonant X-ray scattering in $Ce_{0.7}La_{0.3}B_6$ Evidence for octupole order

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The azimuthal angle dependence observed in the resonant X-ray scattering in phase IV of $Ce_{0.7}La_{0.3}B_6$ is analyzed theoretically [1].

On the basis of the compact formalism for the resonant X-ray scattering [2], it is shown that the peculiar angle dependence observed in the E2 channel is consistent with the Γ_{5u}-type octupole order with principal axis along [111] and equivalent directions. Under the assumption that the four equivalent octupole domains are nearly equally populated in the sample, the observed angle dependences are reproduced by calculation for both σ-σ' and σ-π' polarizations as shown in Fig. 1. In the non-rotated σ-σ' scattering channel, only the Γ_{5u}-type octupole in three-fold principal axis yields consistent six-fold oscillatory behavior with the maximum at $\psi = 0$. On the other hand, in the rotated σ-π' channel, the contribution from the other three equivalent domains predominates over the contribution from the [111] domain, showing three-fold oscillation with the maximum at $\psi = 0$.

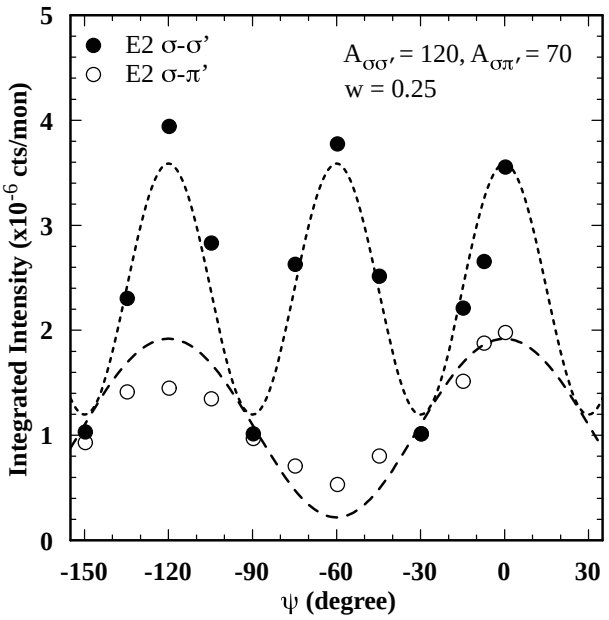

Figure 1: The azimuthal angle dependence. The symbols are taken from Mannix et al. [3]. The dotted and the dashed lines show the calculated intensities [1] from the Γ_{5u}-type octupole order.

The calculation for various symmetries of order parameters excludes unambiguously other order parameters than the Γ_{5u}-type octupole.

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Electronic states near quantum critical point of valence transition

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In $CeCu_2Ge_2$ and $CeCu_2Si_2$, it has been observed that the superconducting(SC) transition temperature takes maximum in the regime where valence change is prominent under pressure, and the relation between valence fluctuation and the mechanism of the superconductivity attracts much attention [1,2]. The similar behavior has been observed in the other Ce compounds such as $CeCoIn_5$ [3] and in $CeIrIn_5$ the SC transition temperature increases in spite that the antiferromagnetic(AF) fluctuation is suppressed under pressure, which suggests a new SC mechanism different from the AF-fluctuation mediated one [4].

To get insight into the valence instability in the Ce systems, the ground-state properties of the periodic Anderson model with the Coulomb repulsion between f and conduction electrons are studied by using the density matrix renormalization group method. The ground-state phase diagram is accurately determined and it is found that the first-order valence transition takes place with the quantum critical point(QCP). It is also found that the SC correlation becomes dominant near the QCP of the valence transition. Critical nature of the valence transition and the effect of the valence instability on the superconductivity will be discussed in detail in the presentation.

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