

平成 27~31 年度 文部科学省 科学研究費補助金 新学術領域研究(研究領域提案型)領域番号: 2704

J-Physics:多極子伝導系の物理



平成 29 年度領域全体会議

平成 30 年 3 月 15 日 (木) 13:00 ~ 3 月 17 日 (土) 15:50 東京大学物性研究所 6 F 大講議室 (千葉県柏市柏の葉 5 丁目 1-5)



3月15日(木)

### オープニング

13:00 - 13:15	播磨尚朝(神戸大理)	はじめに
A01 計画研究		座長:中辻 知
13:15 - 14:40	中辻 知(東大物性研) 中西 良樹(岩手大理工)	局在多極子と伝導電子の相関による新しい量子現象 超音波で探る PrV <sub>2</sub> Al <sub>20</sub> の低温秩序相と電子状態
	小林 寿夫(兵県大物質)	YbAlB4 の低温・高圧力下 <sup>174</sup> Yb 放射光メスバウアー分光法に よる研究
	榊原 俊郎(東大物性研)	EuPtSiの磁気相図
	佐藤 卓(東北大多元研)	Nonreciprocal magnons in noncentrosymmetric magnets
	鈴木 通人(理研 CEMS)	クラスター多極子による反強磁性相の巨視的秩序パラメータ
	有田 亮太郎 (理研 CEMS)	Mn <sub>3</sub> Sn における大きな磁気光学カー効果に対するクラスター 多極子理論

14:40-15:00 休憩

A01 公募研究1

15:00 - 15:20	出口 和彦(名大院理)	正 20 面体準結晶の磁性と超伝導
15:20 - 15:40	渡辺 真仁(九工大基礎科学)	Yb 系準結晶と近似結晶における新しい量子臨界現象
15:40 - 16:00	関山 明(阪大基礎工)	角度分解内殻光電子線二色性による占有 4f 軌道対称性の 解明
16:00 - 16:20	古賀 昌久(東工大理)	ハニカム格子上多軌道模型におけるスピン軌道相互作用 の効果

16:20-16:40 休憩

A01 公募研究 2

#### 座長:中西 良樹

16:40 - 17:00	椎名 亮輔(琉球大理)	Sm 化合物における軌道依存型混成による秩序化現象と重い 電子状態
17:00 - 17:20	大槻 純也(東北大理)	量子多体物理におけるスパースモデリングの方法
17:20 - 17:40	坂井 徹 (兵庫県立大物質理)	低次元量子反強磁性体のスピンネマティック相
17:40 - 18:00	森 道康(原子力機構先端研)	拡張多極子によるフォノンホール効果

### 3月16日(金)

招待講演			座長:柳瀬 陽一
9:15 - 9:55	伊藤 哲明(	(東京理科大理)	単体 Te における電流誘起バルク磁性
9:55 - 10:15	休憩		
C01 計画研究			座長:網塚 浩
10:15 - 11:30	網塚 浩(	(北大理)	反強磁性金属における電気磁気効果の検証
	藤秀樹(	(神戸大理)	UNi₄B の常磁性およびトロイダル磁気秩序状態の B-NMR に よる研究
	中尾 裕則(	(KEK-IMSS)	Resonant x-ray scattering study on hybridized orbital states in d- and f-electron system
	高阪 勇輔(	(岡大基礎研)	無機キラル磁性体の不斉結晶育成
	楠瀬 博明(	(明大理工)	拡張多極子による交差相関結合
	御領 潤(	(弘前大理工)	Kane-Mele 金属におけるカイラル d-波超伝導と表面自発ス ピン分極: SrPtAs を視野に入れて
C01 公募研究 1			座長:藤 秀樹
11:30 - 11:50	大串 研也(	(東北大理)	奇パリティ多極子秩序伝導系の開拓
11:50 - 12:10	大原 繁男(	(名工大院工)	反転中心のない結晶構造の YbNi₃Al₀ および関連物質の磁気 的性質
12:10 - 12:30	高畠 敏郎 (	広大院先端物質)	ジグザグ鎖近藤半導体の反強磁性秩序に対する一軸圧力効果
12:30 - 13:40	記念撮影·	昼食	
13:40			
	ポスターセ	ッション	
16:10			
C01 公草研空 2			広 に い は 祖 田
1(.10 1(.00	日四	(达上田)	
16:10 - 16:30 16:30 - 16:50	而回 見 古賀 幹人	(坷入理) (静大教育) ;	動的平均場近似に基つく弗一原理訂昇手法の開発と応用 結晶場多重項に起因する非フェルミ流体状態における多極子物理

16:50 - 16:55	小休憩
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D01	計画研究

## 座長:野原 実

16:55 - 18:00	野原 実(岡大基礎研)	5d 遷移金属を含む超伝導体の開発
	鬼丸 孝博(広大院先端研)	Pr 系の非クラマース二重項における相転移と非フェルミ液体 的挙動
	水口 佳一(首都大物理)	重金属を含む層状超伝導体の探索
	秋光 純 (岡大基礎研)	Trial to Carrier-doping in Sr <sub>2</sub> IrO <sub>4</sub> /Ba <sub>2</sub> IrO <sub>4</sub>
	播磨 尚朝(神戸大理)	反転対称の無い系やウラン化合物の電子構造

18:30-20:30 懇談会(東大物性研究所カフェテリア)

### 3月17日(土)

D01 公募研究		座長:水口 佳一
9:30 - 9:50	高林 康裕 (東北大 AIMR)	Emergent electronic phenomena in hybrid f-/p-electron molecular materials
9:50 - 10:10	阿部 伸行(東大新領域)	3d 電子系における高次多極子に由来する非対角応答
10:10 - 10:30	岡本 佳比古(名大院工)	Ir・Pt 化合物における新超伝導体探索
10:30 - 10:50	宮坂 茂樹(阪大院理)	遷移金属ダイカルコゲナイドの異常物性

10:50-11:10 休憩

# B01 計画研究

座長:青木 大

青木 大(東北大金研)	B01 計画研究のこれまでの成果と f 電子系化合物の超伝導探索
石田 憲二(京大理)	重い電子人工超格子の核磁気共鳴
神戸 振作 (原子力機構先端研)	URu <sub>2</sub> Si <sub>2</sub> のNMRによる研究
井澤 公一(東工大理)	U <sub>1-x</sub> Th <sub>x</sub> Be <sub>13</sub> の輸送係数にみられる非フェルミ液体的挙動
松田 達磨(首都大院理工)	SmTr <sub>2</sub> Al <sub>20</sub> (Tr:遷移金属) における重い電子状態の特徴について
柳瀬 陽一(京大理)	奇パリティ多極子秩序・超伝導の分類学と電磁応答
	青木       大(東北大金研)         石田       憲二(京大理)         神戸       振作(原子力機構先端研)         井澤       公一(東工大理)         松田       達磨(首都大院理工)         柳瀬       陽一(京大理)

12:25-13:35 昼食

### B01 公募研究

座長:石田 憲二

13:35 - 13:55	服部 一匡(首都大院理工)	異方的四極子 RKKY 相互作用と磁場中強四極子秩序
13:55 - 14:15	池田 浩章(立命館理工)	多軌道超伝導体の分類とその応用
14:15 - 14:35	藤森 伸一(原子力機構)	3 次元 ARPES による強相関ウラン化合物の電子状態
14:35 - 14:55	野尻 浩之(東北大金研)	URu <sub>2</sub> Si <sub>2</sub> および関連化合物の強磁場相
14:55 - 15:15	野島 勉(東北大金研)	電場誘起超伝導体における増強された常磁性極限臨界磁場の 直接観測
15:15 - 15:35	大貫 惇睦(琉大理)	立方晶化合物の特異なフェルミ面

クロージング

15:35 – 15:50 評価委員コメント 領域代表挨拶

# ポスターセッション (3月16日 13:40-16:10)

P01:	金杉	翔太	京大理	Spin-Orbit Coupled Ferroelectric Superconductor
P02:	石塚	淳	京大理	局所反転対称性のない系における奇パリティ多極子揺らぎと超伝導
P03:	本山	岳	島根大総合理	新しい重い電子系化合物 Ce <sub>3</sub> TiBi <sub>5</sub> の物性測定
P04:	坂本	直樹	京大理	URu2Si2の超伝導相におけるトポロジカル端状態
P05:	梅尾	和則	広大自然セ	12 GPa までの交流法比熱測定によるキラル磁性体 YbNi <sub>3</sub> Ga <sub>9</sub> の圧力誘起 磁気秩序相の研究
P06:	齋藤	開	北大理	CeRu <sub>2</sub> Al <sub>10</sub> における電流誘起磁化現象
P07:	水戸	毅	兵庫県大院物質理	Sm 化合物で観測される特異な 4f 電子二重性
P08:	三宅	和正	阪大先端強磁場	Ce <sub>x</sub> La <sub>1-x</sub> Cu <sub>5.62</sub> Au <sub>0.38</sub> (x = 0.02 – 0.10) で観測された非従来型非フェルミ 液体的振る舞いの理論
P09:	谷口	貴紀	東大物性研	四極子秩序相を持つ PrTi <sub>2</sub> Al <sub>20</sub> の異方的強四極子間相互作用: Al-NMR と磁化による研究
P10:	池田	暁彦	東大物性研	コバルト酸化物におけるスピンと軌道の多極子秩序
P11:	松田	康弘	東大物性研	YbB12の c-f 混成ギャップの磁場制御と磁場誘起一近藤金属状態
P12:	角田	峻太郎	京大理	角運動量 jz に依存する超伝導ポイントノード — UPt3, UBe13, SrPtAs などへの適用—
P13:	富田	崇弘	東大物性研	ワイル磁性体 Mn <sub>3</sub> Sn における異常ホール効果と磁化
P14:	Qu Da	anru	東大物性研	Inverse spin Hall effect in Mn-Sn amorphous alloy thin film
P15:	真砂	全宏	京大院理	強磁性超伝導体 UCoGe の圧力下超伝導の研究
P16:	日高	宏之	北大理	局所空間反転対称性の破れた SmBe <sub>13</sub> の磁気構造に関する研究
P17:	渡邊	光	京大理	反強磁性体ドメインスイッチングに関する対称性の考察
P18:	大岩	陸人	明大理工	ホールドープした単層 MoS2 における超伝導の理論
P19:	速水	賢	北大理	Microscopic Description of Electric and Magnetic Toroidal Multipoles in Hybrid Orbitals
P20:	和田	泰地	名大院工	1 次元テルル化物 M <sub>4</sub> SiTe <sub>4</sub> (M = Ta, Nb)の熱電特性
P21:	北川	俊作	京大理	FFLO state in CeCu <sub>2</sub> Si <sub>2</sub> revealed by Cu-NMR
P22:	鬼丸	孝博	広大院先端研	Pr1-2-20 系における非フェルミ液体的挙動
P23:	常盤	欣文	アウグスブルク大	PrIr <sub>2</sub> Zn <sub>20</sub> における四極子臨界磁場近傍での異常な熱膨張
P24:	大槻	匠	東大物性研	パイロクロア型 Pr <sub>2</sub> Ir <sub>2</sub> O <sub>7</sub> 薄膜における磁気抵抗の磁場・角度依存性
P25:	柳澤	達也	北大理	パルス強磁場下超音波測定による URu2Si2 における多極子不安定性の探索
P26:	舩島	洋紀	神戸大院理	軌道近藤効果に適したバンド構造と実例
P27:	平井	大悟郎	東大物性研	スピン軌道結合金属 Cd₂Re₂O7 におけるスピン分裂した Fermi 面の観測

P28:	肥後	友也	東大物性研	Magnetic octupole induced large magneto-optical Kerr effect in the AF Weyl metal $Mn_3Sn$
P29:	植木	輝	北大理	s 波超伝導体の孤立渦における渦糸フローホール効果の理論
P30:	松林	和幸	電通大	PrT <sub>2</sub> Al <sub>20</sub> (T = Ti, V)の高圧下における四極子秩序と超伝導
P31:	酒井	明人	東大物性研	Anomalous Hall effect and magnetoresistance in the nodal metallic spin ice $Pr_2Ir_2O_7$
P32:	呉	昌根	岡大基礎研	Time-dependent reentrant superconductivity in nonequilibrium $\text{KBi}_2$
P33:	工藤	一貴	岡大基礎研	Low temperature synthesis and specific heat study of Chevrel phase compounds $Mo_6Ch_8$ (Ch = S, Se, Te)
P34:	工藤	一貴	岡大基礎研	Superconductivity in $Mg_2Ir_3Si$ : an ordered variant of the hexagonal Laves phase $MgZn_2$
P35:	後藤	陽介	首都大	SnAs 層を含む新しい層状超伝導体 NaSn <sub>2</sub> As <sub>2</sub> と元素置換効果
P36:	唐	楠	東大物性研	Temperature and Field Dependence of Lattice Elasticity in $% 10^{-10}$ Quantum Spin Ice Material $Pr_{2}Zr_{2}O_{7}$
P37:	星	和久	首都大物理	BiCh <sub>2</sub> 系超伝導体 LaO <sub>0.6</sub> F <sub>0.4</sub> Bi(S,Se) <sub>2</sub> の Se 同位体効果
P38:	曽我福	部 遼太	首都大物理	Ce を含む BiS2系超伝導体の新規合成
P39:	小林	達生	岡大自然科学	Cd <sub>2</sub> Re <sub>2</sub> O <sub>7</sub> の高圧下ホール効果
P40:	橘高	俊一郎	東大物性研	Field-orientation dependence of a ferroquadrupole order in $PrTi_2Al_{20}$ investigated by thermal measurements
P41:	井倉	拓哉	岡大基礎研	Superconductivity in $IrIn_2$ with iridium infinite chains: A comparative study of $CoIn_2$ and $IrIn_2$
P42:	Chen	Taishi	東大物性研	Intrinsic Origin Investigation on AHE and Thermoelectric Behavior
P43:	八城	愛美	北大理	Odd-parity multipoles by orbital hybridization in noncentrosymmetric tetragonal system
P44:	鈴木	慎太郎	東大物性研	価数揺動系 <i>α</i> -YbAlB <sub>4</sub> の異常金属状態に対する非弾性中性子散乱研究
P45:	荒木	幸治	防衛大応物	六方晶 Yb <sub>1-x</sub> Lu <sub>x</sub> CuGe の磁性
P46:	野間	雄一朗	神戸大理	強磁性超伝導体 UGe2の圧力下 <sup>73</sup> Ge-NQR による磁気ゆらぎの研究
P47:	永岡	靖浩	東大物性研	低温熱膨張および磁歪測定による四極子近藤格子 PrV <sub>2</sub> Al <sub>20</sub> における 多極子秩序の研究
P48:	堀江	理恵	岡大異分野基礎研	Superconductivity in the laves phase SrIr <sub>2</sub>
P49:	柳瀬	陽一	京大理	UPt3 における非共型トポロジカル超伝導
P50:	小林	夏野	岡大基礎研	化学ドープによる binary chalcogenides の超伝導転移温度上昇
P51:	中村	直貴	首都大理工	遍歴弱強磁性体充填スクッテルダイト化合物 LaFe <sub>4</sub> As <sub>12</sub> の高純度単結晶 育成と dHvA 効果
P52:	大村	瑠美	首都大理工	大きな単位胞を取る三元化合物 <i>R</i> -Ir-Sn( <i>R</i> :希土類)の単結晶育成と 構造解析
P53:	柳	有起	明大理工	スレーブボソン平均場近似による多軌道周期アンダーソン模型の解析

P54:	宮脇 瑠美佳	首都大理工	四極子近藤格子系 PrTr <sub>2</sub> Al <sub>20</sub> (Tr = Mo, W)の単結晶育成と低温物性測定
P55:	Ikhlas Muhammad	東大物性研	The Effect of Doping on the Transport Properties of the Non-collinear Antiferromagnet $Mn_3Sn$
P56:	小手川 亘	神戸大理	GaTa4Se8のNMR 測定
P57:	仲嶺 元輝	京大理	NMR による人工超格子 CeCoIn5/CeRhIn5の研究
P58:	松井 隆志	神戸大院理	NMR によるテトラヘドライト Cu <sub>12</sub> Sb <sub>4</sub> S <sub>13</sub> におけるダイナミクスの研究
P59:	藤原 賢二	島根大院総理工	重い電子系超伝導体 CeCu <sub>2</sub> Si <sub>2</sub> の高圧・強磁場下の NMR
P60:	仲村 愛	東北大金研	単結晶 YbNiSn の磁性と磁気相図
P61:	田端 千紘	KEK IMSS	トロイダル秩序物質 UNi4B の単結晶 X 線構造解析
P62:	Maurya Arvind	東北大金研	反転対称性を持たない単結晶 URhSn の圧力下の磁気相転移
P63:	西川 尚	東大物性研	ワイル磁性体薄膜の熱電効果
P64:	久保 徹郎	岡山理大理	NMR/NQR Spectrum Analysis and High-Field Magnetization in $PrT_2Al_{20}$ ( $T = Nb, Ta$ )

FY2015~2019 MEXT KAKENHI on Innovative Areas #2704

# J-Physics : Physics of Conductive Multipole Systems



FY 2017 Annual Meeting Thu. March 15, 13:00 ~ Sat. March 17, 15:50 Large Meeting Room, 6F, The Institute of Solid State Physics

The University of Tokyo (5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581 Japan)



Chair: Yoshiki Nakanishi

科研

### March 15 (Thu.)

#### Opening

13:00 - 13:15	Hisatomo Harima
	Kobe University

#### **Opening Remarks**

#### A01 Planned Research Chair: Satoru Nakatsuji 13:15 - 14:40Satoru Nakatsuji Novel quantum phenomena induced by the correlation between localized multipoles and conduction electrons The University of Tokyo Yoshiki Nakanishi Low-Temperature Elastic Properties of PrV2Al20 probed by Iwate University ultrasonic measurements Synchrotron radiation based <sup>174</sup>Yb Mössbauer spectroscopic study Hisao Kobayashi of YbAlB4 under pressure at low temperature The University of Hyogo Magnetic Phase Diagram of EuPtSi Toshiro Sakakibara The University of Tokyo Taku J. Sato Nonreciprocal magnons in noncentrosymmetric magnets Tohoku University Michi-To Suzuki Macroscopic order parameters for antiferromagnetic phases by RIKEN AICS cluster multipole moments Rvotaro Arita Cluster multipole theory for large magneto-optical Kerr effect in **RİKEN AICS** Mn<sub>3</sub>Sn

14:40 - 15:00Coffee Break

### A01 Invited Research 1

A01 Invited Res	earch 1	Chair: Toshiro Sakakibara
15:00 - 15:20	Kazuhiko Deguchi Nagoya University	Magnetism and Superconductivity of Icosahedral Quasicrystals
15:20 - 15:40	Shinji Watanabe Kyushu Institute of Technology	New quantum criticality in Yb-based quasicrystal and approximant crystal
15:40 - 16:00	Akira Sekiyama Osaka University	Probing occupied 4f-orbital symmetry by linear dichroism in angle- resolved core-level photoemission
16:00 - 16:20	Akihisa Koga Tokyo Institute of Technology	Role of the spin-orbit coupling in the Kugel-Khomskii model on the honeycomb lattice
16:20 - 16:40	Coffee Break	

### A01 Invited Research 2

16:40 - 17:00	Ryousuke Shiina University of the Ryukyus	Ordering and fermi-liquid Properties due to orbital-dependent hybridization in Sm compounds
17:00 - 17:20	Junya Otsuki Tohoku University	Methods of sparse modeling in quantum many-body physics
17:20 - 17:40	Toru Sakai The University of Hyogo	Spin Nematic Physics in Low-Dimensional Quantum Antiferromagnets
17:40 - 18:00	Michiyasu Mori Japan Atomic Energy Agency	Phonon Hall effect by extended cluster multipoles

March 16 (Fri.)		
Invited Talk		Chair: Youichi Yanase
9:10 - 9:55	Tetsuaki Itou Tokyo University of Science	Current-induced bulk magnetization in elemental tellurium
9:55 - 10:15	Coffee Break	
C01 Planned Re	esearch	Chair: Hiroshi Amitsuka
10:15 - 11:30	Hiroshi Amitsuka Hokkaido University	Tests for Magnetoelectric Effects on Antiferromagnetic Metals
	Hideki Tou Kobe University	B-NMR study on paramagnetic and toroidal magnetic ordered states in UNi <sub>4</sub> B
	Hironori Nakao <i>KEK IMSS</i>	Resonant x-ray scattering study on hybridized orbital states in d- and f- electron system
	Yusuke Kousaka Hiroshima University	Homo-chiral crystallization in inorganic chiral magnetic materials
	Hiroaki Kusunose Meiji University	Cross-correlated coupling based on augmented multipoles
	Jun Goryo Hirosaki University	Surface properties of a chiral d-wave superconductor with hexagonal symmetry
C01 Invited Research		Chair: Hideki Tou
11:30 - 11:50	Kenya Ohgushi Tohoku University	Exploration of materials with odd-parity multipole order
11:50 - 12:10	Shigeo Ohara Nagoya Institute of Technology	Magnetic properties of YbNi <sub>3</sub> Al <sub>9</sub> and related materials with a non- centrosymmetric crystal structure
12:10 - 12:30	Toshiro Takabatake Hiroshima University	Uniaxial pressure effect on the antiferromagnetic order in the Kondo semiconductors with zigzag chains
12:30 - 13:40	Group Photo • Lunch	
13:40		
	Poster Session	

16:10

### **C01 Invited Research**

C01 Invited Res	earch	Chair: Hiroaki Kusunose
16:10 - 16:30	Hiroshi Shinaoka Saitama University	Development of <i>ab-inito</i> method based on dynamical mean- field approximation and applications
16:30 - 16:50	Mikito Koga Shizuoka University	Multipole physics in non-Fermi-liquid state due to a crystal-field multiplet
16:50 - 16:55	Short Break	

# **D01 Planned Research**

D01 Planned Research		Chair: Minoru Nohara
16:55 - 18:00	Minoru Nohara Okayama University	Exploration of novel superconductors with 5d transition metal
	Takahiro Onimaru Hiroshima University	Two-channel Kondo Behaviors and Unconventional Phase transitions in Pr-based non-Kramers doublet systems
	Yoshikazu Mizuguchi Tokyo Metropolitan University	Exploration for new layered superconductors with heavy elements
	Jun Akimitsu Okayama University	Trial to Carrier-doping in Sr <sub>2</sub> IrO <sub>4</sub> /Ba <sub>2</sub> IrO <sub>4</sub>
	Hisatomo Harima Kobe University	Electronic structure of noncentrosymmetric system and Uranium compounds

Get-Together and Free Discussion (ISSP Cafeteria) 18:30 - 20:30

### March 17 (Sat.)

### **D01 Invited Research**

# Chair: Yoshikazu Mizuguchi

9:30 - 9:50	Yasuhiro Takabayashi Tohoku University	Emergent electronic phenomena in hybrid f-/p-electron molecular materials
9:50 - 10:10	Nobuyuki Abe The University of Tokyo	Odd parity multipole ordering and off-diagonal response in 3d electron system
10:10 - 10:30	Yoshihiko Okamoto Nagoya University	Search for New Ir- and Pt-Based Superconductors
10:30 - 10:50	Shigeki Miyasaka Osaka University	Anomalous Electronic Properties in Transition Metal Dichalcogenides

10:50 - 11:10 Coffee Break

# **B01 Planned Research**

301 Planned Research		Chair: Dai Aoki
11:10 - 12:25	Dai Aoki Tohoku University	Results of B01 group and search for superconductivity in f-electron systems
	Kenji Ishida Kyoto University	NMR study on the heavy-fermion superlattices
	Shinsaku Kambe Japan Atomic Energy Agency	NMR study of URu <sub>2</sub> Si <sub>2</sub>
	Koichi Izawa Tokyo Institute of Technology	Non Fermi liquid behavior in transport coefficients of $U_{1-x}Th_xBe_{13}$
	Tatsuma D. Matsuda Tokyo Metropolitan University	Characteristic Heavy Electron State in Sm <i>Tr</i> <sub>2</sub> Al <sub>20</sub> ( <i>Tr</i> : transition metal)
	Youichi Yanase Kyoto University	Classification theory of odd-parity multipole order/superconductivity and electromagnetic responses

12:25 - 13:35 Lunch

### **B01 Invited Research**

B01 Invited Res	earch	Chair: Kenji Ishida
13:35 - 13:55	Kazumasa Hattori Tokyo Metropolitan University	Anisotropic quadrupole RKKY interactions under magnetic fields: ferro quadrupole order in PrTi <sub>2</sub> Al <sub>20</sub>
13:55 - 14:15	Hiroaki Ikeda Ritsumeikan University	Classification of multi-orbital superconductivity and its application
14:15 - 14:35	Shin-ichi Fujimori Japan Atomic Energy Agency	Electronic structures of strongly correlated uranium compounds studied by three-dimensional ARPES
14:35 - 14:55	Hiroyuki Nojiri Tohoku University	High Magnetic Field Phases of $URu_2Si_2$ and Related Compounds
14:55 - 15:15	Tsutomu Nojima Tohoku University	Direct Observation of Enhanced Paramagnetic-Limited Critical Magnetic Field in Electric-Field-Induced Superconductors
15:15 - 15:35	Yoshichika Ōnuki University of the Ryukyus	Unique Fermi Surface Properties in Cubic Compounds

# Closing

15:35 - 15:50	Advisors' Comments
	Closing Remarks by Area Representative

# Poster Session (March 16, 13:40 - 16:10)

P01:	Shota Kanasugi Kyoto University	Spin-Orbit Coupled Ferroelectric Superconductor
P02:	Jun Ishizuka Kyoto University	Odd-parity multipole fluctuation and superconductivity in locally noncentro- symmetric system
P03:	Gaku Motoyama Shimane University	Magnetic properties of the new heavy fermion compounds Ce3TiBi5
P04:	Naoki Sakamoto Kyoto University	The topological edge state of $URu_2Si_2$ in superconducting state
P05:	Kazunori Umeo Hiroshima University	Pressure-induced magnetic phase in the chiral magnet YbNi <sub>3</sub> Ga <sub>9</sub> : AC-calorimetric measurements up to 12 GPa
P06:	Hiraku Saito Hokkaido University	Current-induced magnetization on CeRu <sub>2</sub> Al <sub>10</sub>
P07:	Takeshi Mito The University of Hyogo	Novel dual character of 4f electrons observed in Sm compounds
P08:	Kazumasa Miyake Osaka University	Theory for Unconventional Non-Fermi Liquid Behaviors Observed in $Ce_xLa_{1-x}Cu_{5.62}Au_{0.38}$ ( $x = 0.02 - 0.10$ )
P09:	Takaki Taniguchi The University of Tokyo	Anisotropic ferro-quadupole interactions in quadrupole ordered system $PrTi_2Al_{20}$ : Al-NMR and Magnetization Studies
P10:	Akihiko Ikeda The University of Tokyo	Multipole order of spin and orbitals in cobaltites
P11:	Yasuhiro H. Matsuda The University of Tokyo	Magnetic-field-induced Kondo metal state by closing the c-f hybridization gap in $YbB_{12}$
P12:	Shuntaro Sumita Kyoto University	Superconducting point nodes depending on angular momentum $j_z$ —Application to UPt <sub>3</sub> , UBe <sub>13</sub> , SrPtAs, etc.—
P13:	Takahiro Tomita The University of Tokyo	Anomalous Hall effect and magnetization in Weyl Magnet Mn <sub>3</sub> Sn
P14:	<b>Qu Danru</b> The University of Tokyo	Inverse spin Hall effect in Mn-Sn amorphous alloy thin film
P15:	Masahiro Manago Tokyo Institute of Technology	Study of the superconductivity of the ferromagnetic superconductor UCoGe under pressure
P16:	Hiroyuki Hidaka Hokkaido University	Study of Magnetic Structure in SmBe <sub>13</sub> with broken local-inversion symmetry
P17:	Hikaru Watanabe Kyoto University	Symmetry analysis of electrical switching of antiferromagnet
P18:	Rikuto Oiwa Meiji University	Theory of superconductivity in hole-doped monolayer $MoS_2$
P19:	Satoru Hayami Hokkaido University	Microscopic Description of Electric and Magnetic Toroidal Multipoles in Hybrid Orbitals
P20:	Taichi Wada Nagoya University	Thermoelectric properties of one dimensional telluride $M_4$ SiTe <sub>4</sub> ( $M$ = Ta, Nb)
P21:	Shunsaku Kitagawa Kyoto University	FFLO state in CeCu <sub>2</sub> Si <sub>2</sub> revealed by Cu-NMR
P22:	Takahiro Onimaru Hiroshima University	Non-Fermi Liquid Behaviors in Pr 1-2-20 Systems
P23:	Yoshifumi Tokiwa University of Augsburg	Anomalous thermal expansion near the quadrupolar critical field in $\mbox{Pr}\mbox{Ir}_2\mbox{Zn}_{20}$
P24:	Takumi Ohtsuki The University of Tokyo	Field and angular dependent magnetoresistance in $Pr_2Ir_2O_7$ thin films
P25:	Tatsuya Yanagisawa Hokkaido University	Search for Multipolar Instability in $URu_2Si_2$ Studied by Ultrasonic Measurements under Pulsed Magnetic Field

P26:	Hiroki Funashima Kobe University	Electronic structures suitable for the orbital Kondo effect and actual materials
P27:	Daigoro Hirai The University of Tokyo	Observation of spin-split Fermi surfaces in the spin-orbit coupled metal Cd <sub>2</sub> Re <sub>2</sub> O <sub>7</sub>
P28:	Tomoya Higo The University of Tokyo	Magnetic octupole induced large magneto-optical Kerr effect in the AF Weyl metal $Mn_3 Sn$
P29:	Hikaru Ueki Hokkaido University	Theory of the flux-flow Hall effect in an isolated vortex of an s-wave superconductor
P30:	Kazuyuki Matsubayashi The University of Electro- Communications	High pressure studies of quadrupole order and superconductivity in $PrT_2Al_{20}$ (T = Ti, V)
P31:	Akito Sakai The University of Tokyo	Anomalous Hall effect and magnetoresistance in the nodal metallic spin ice Pr <sub>2</sub> Ir <sub>2</sub> O <sub>7</sub>
P32:	Chang-geun Oh Okayama University	Time-dependent reentrant superconductivity in nonequilibrium $\mathrm{KBi}_2$
P33:	Kazutaka Kudo Okayama University	Low temperature synthesis and specific heat study of Chevrel phase compounds $Mo_6Ch_8$ (Ch = S, Se, Te)
P34:	Kazutaka Kudo Okayama University	Superconductivity in $Mg_2Ir_3Si:$ an ordered variant of the hexagonal Laves phase $MgZn_2$
P35:	Yosuke Goto Tokyo Metropolitan University	Effect of elemental substitution for SnAs-based novel layered superconductor $NaSn_2As_2$
P36:	Nan Tang The University of Tokyo	Temperature and Field Dependence of Lattice Elasticity in Quantum Spin Ice Material $Pr_2Zr_2O_7$
P37:	Kazuhisa Hoshi Tokyo Metropolitan University	Se Isotope Effect in $LaO_{0.6}F_{0.4}Bi(S,Se)_2$ Superconductor
P38:	Ryota Sogabe Tokyo Metropolitan University	Synthesis of Bi-chalcogenide superconductor with Ce-based blocking layer
P39:	Tatsuo C. Kobayashi Okayama University	Hall effect in Cd <sub>2</sub> Re <sub>2</sub> O <sub>7</sub> under high pressure
P40:	Shunichiro Kittaka The University of Tokyo	Field-orientation dependence of a ferroquadrupole order in $\mbox{PrTi}_2\mbox{Al}_{20}$ investigated by thermal measurements
P41:	Takuya Ikura Okayama University	Superconductivity in $IrIn_2$ with iridium infinite chains: A comparative study of $CoIn_2$ and $IrIn_2$
P42:	Chen Taishi The University of Tokyo	Intrinsic Origin Investigation on AHE and Thermoelectric Behavior
P43:	Megumi Yatsushiro Hokkaido University	Odd-parity multipoles by orbital hybridization in noncentrosymmetric tetragonal systems
P44:	Shintaro Suzuki The University of Tokyo	The inelastic neutron scattering study on the strange metal behavior in the intermediate valence system $\alpha$ -YbAlB <sub>4</sub>
P45:	Koji Araki National Defense Academy	Magnetic properties of Hexagonal Yb <sub>1-x</sub> Lu <sub>x</sub> CuGe
P46:	Yuichiro Noma Kobe University	$^{73}\mbox{Ge-NQR}$ studies under pressure on magnetic fluctuations of ferromagnetic superconductor $UGe_2$
P47:	Yasuhiro Nagaoka The University of Tokyo	Studies of multipole order in quadrupole Kondo lattice system $PrV_2Al_{20}$ by thermal expansion and magnetostriction
P48:	Rie Horie Okayama University	Superconductivity in the laves phase SrIr <sub>2</sub>
P49:	Youichi Yanase Kyoto University	Nonsymmorphic topological superconductivity in UPt <sub>3</sub>
P50:	Kaya Kobayashi Okayama University	Enhancement of $T_c$ in binary chalcogenides by chemical doping

P51:	Naoki Nakamura Tokyo Metropolitan University	de Hass-van Alphen effect of the itinerant weak ferromagnetic filled skutterudite $LaFe_4As_{12}$
P52:	Rumi Omura Tokyo Metropolitan University	Single crystal growth and structural analysis of ternary compound $R$ -Ir-Sn ( $R$ : rare earth) with large unit cell
P53:	Yuki Yanagi Meiji University	Slave boson mean field study of multi-orbital periodic Anderson model
P54:	Rumika Miyawaki Tokyo Metropolitan University	Single crystal growth and low-temperature physical property measurements of quadrupolar Kondo lattice $PrTr_2Al_{20}$ ( $Tr = Mo, W$ )
P55:	Ikhlas Muhammad The University of Tokyo	The Effect of Doping on the Transport Properties of the Non-collinear Antiferromagnet $Mn_3Sn$
P56:	Hisashi Kotegawa Kobe University	NMR Study of GaTa <sub>4</sub> Se <sub>8</sub>
P57:	Genki Nakamine Kyoto University	NMR study on the artificial superlattice CeCoIn <sub>5</sub> /CeRhIn <sub>5</sub>
P58:	Takashi Matsui Kyoto University	NMR study on dynamical property of tetrahedrite $Cu_{12}Sb_4S_{13}$
P59:	Kenji Fujiwara Shimane University	NMR study of CeCu <sub>2</sub> Si <sub>2</sub> under high pressure and field
P60:	Ai Nakamura Tohoku University	Magnetic Properties and Magnetic Phase Diagram of Single Crystalline YbNiSn
P61:	Chihiro Tabata KEK IMSS	X-ray Crystal Structure Analysis of Single-Crystalline UNi <sub>4</sub> B
P62:	Maurya Arvind Tohoku University	Magnetic Phase Transitions in Single Crystalline Non- centrosymmetric URhSn under Pressure
P63:	Takashi Nishikawa	Thermoelectric properties of a Weyl magnet thin film
P64:	Tetsuro Kubo Okayama University of Science	NMR/NQR Spectrum Analysis and High-Field Magnetization in $PrT_2Al_{20}$ ( $T = Nb$ , Ta)

# Novel quantum phenomena induced by the correlation between localized multipoles and conduction electrons

### Satoru Nakatusji<sup>1</sup>

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The subject of A01 group is to study the novel quantum phenomena driven by the strong hybridization between localized mulitipoles and conduction electrons. I will first review the recent highlights in the results made by the members in the A01 group. Then, I will talk about our recent work on the complex multipolar degrees of freedom such as valence and quadrupolar degrees of freedom, which induces anomalous metallic states and novel superconductivity. We will discuss interesting cases found using the intermediate valence systems YbAlB<sub>4</sub> and quadrupolar Kondo systems PrTr<sub>2</sub>Al<sub>20</sub> [1-3]. If time permits, I will talk about novel phenomena induced by itinerant cluster magnetic octuple found in the antiferromagnetic Weyl metal Mn<sub>3</sub>Sn and Mn<sub>3</sub>Ge [4-8].

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# Field-induced quantum criticality in PrV<sub>2</sub>Al<sub>20</sub> proved by ultrasonic measurements

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Heavy fermion quantum criticality is one of the central issues to be solved in contemporary condensed matter physics. It has been recognized that the important examples are seen frequently in metals containing lanthanide atoms with the Frank-Kasper polyhedral type cage structure. They exhibit novel phenomena at low temperatures, like unconventional type of anomalous Hall effect, spin liquid like behaviour, non-Fermi liquid behaviour driven by the hybridization between conduction electrons and quadrupole (orbital) moments, and so on. It was found that correlation between conduction electrons and electronic quadrupole moments, in particular leads to fascinating materials properties. For the Pr-based systems, the  $\Gamma_3$  non-Kramers crystalline-electric-field (CEF) doublet ground state is often realized in a Pr<sup>3+</sup> ion with a 4f<sup>2</sup> configuration under cubic crystalline symmetry. Thus, the Pr-based systems with non-Kramers doublet ground states are suitable for study of the quadrupolar quantum criticality. In  $\Gamma_3$  non-Kramers systems, the local quadrupole moment derived from Pr ion can be scattered by conduction electrons via two equivalent scattering channel ( $O_{20}$ ,  $O_{22}$ ), and the channel frustration sometimes leads to the imperfect screening of the quadrupole moments. Quadrupolar order can be successfully investigated using ultrasonic measurements where elastic anomalies reveal ground state properties including orbital degrees of freedom or motif.

We have investigated elastic properties of the Pr based cage compound  $PrV_2Al_{20}$  (Tr : Ti, V) by means of ultrasonic measurements.  $PrV_2Al_{20}$  has the cubic  $CeCr_2Zn_{20}$ -type structure with the space group Fd3m. Pr and V atoms form a diamond structure and a  $\beta$ -pyrochlore type partial sub-lattices, respectively.  $PrV_2Al_{20}$  exhibits quadrupolar ordering at  $T_Q = 0.65$  K and heavy fermion superconductor with  $T_c = 50$  mK at ambient pressure.[1-2] Furthermore, additional ordering was recently discovered at 0.75 K. We have clearly observed elastic anomalies in the temperature dependence of principal elastic constants at the ordering temperatures of 0.65 K and 0.75 K.[3] Based on the experimental data, magnetic field vs temperature phase diagram was constructed for  $PrV_2Al_{20}$ . The obtained result provides an extended insight into the fundamentals of a quadrupolar ordering derived from the non-Kramers doublet  $\Gamma_3$ . Furthermore, a pronounced elastic softening toward low temperature is revived by applying a magnetic field in the temperature dependence of the elastic constant ( $C_{11}$ - $C_{12}$ )/2, due to the non-Kramers doublet  $\Gamma_3$ , being degenerate in the order parameter space ( $O_{20}$ ,  $O_{22}$ ).[4]

In this talk, we discuss the low temperature elastic property, and also the nature of the order parameter and some types of possible phases in  $PrV_2Al_{20}$ , including a commensurate-incommensurate transition for the quadrupolar moment. In addition, we report the discovery of a field-tuned quantum criticality based solely on the quadrupolar degrees of freedom through the evolution with magnetic fields of the heavy *f*-derived quasiparticles in  $PrV_2Al_{20}$  probed by the ultrasonic measurement.

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- [4] Y. Nakanishi et al, (in preparation)

# Synchrotron radiation based <sup>174</sup>Yb Mössbauer spectroscopic study of YbAlB<sub>4</sub> under pressure at low temperature

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β-YbAlB4 exhibits intrinsic quantum criticalities (QCs) without tuning [1,2], which does not follow the conventional spin-fluctuation theories. The averaged Yb valence in β-YbAlB<sub>4</sub> was found to be intermediate between Yb<sup>2+</sup> and Yb<sup>3+</sup> by means of the hard X-ray photoemission spectroscopic experiments [3], indicating the strong hybridization of the 4*f* electrons of the Yb ions with the conduction electrons. However, the magnetic susceptibilities of β-YbAlB<sub>4</sub> indicate the existences of the localized magnetic moments with Ising anisotropy along the c-axis [1]. Since hydrostatic pressure is an ideal perturbation of the hybridization of *f* electrons with conduction electrons, application of pressure offers a unique possibility to investigate the nature of anomalous QCs in β-YbAlB<sub>4</sub>. Recently, using electrical resistivity measurements under pressure, the Fermi liquid region was observed above ~1 GPa and a magnetic order was suggested above 2.5 GPa at low temperature [4]. However, we need to obtain microscopic information about electronic states of β-YbAlB<sub>4</sub> under pressure to discuss the magnetic order and then we consider the <sup>174</sup>Yb Mössbauer spectroscopic measurements.

The experiments of the synchrotron-radiation (SR)-based <sup>174</sup>Yb ( $E_{\gamma}$ =76.465 keV between the  $I_g$ =0 ground and  $I_e$ =2 excited nuclear states) Mössbauer spectroscopy were carried out using the single crystalline samples under pressures at low temperatures on BL19LXU beamline. The several single-crystal samples approximately 15 µm thick were loaded into the sample cavity of an Inconel alloy gasket with ruby crystals in a clamp-type diamond anvil cell (DAC). To ensure good hydrostatic conditions at low temperatures, we used 4:1 methanol-ethanol as a pressure-transmitting medium. The pressure was calibrated by measuring the wavelength shift of the  $R_1$  luminescence line of the ruby crystals in the clamp-type DAC.

Figure 1 shows the selected SR-based <sup>174</sup>Yb Mössbauer spectrum of  $\beta$ -YbAlB<sub>4</sub> observed at 2 K and 2.45 GPa. As seen in Fig. 1 where the propagation vector of the incident X-ray was parallel to the *c*-axis of the single crystalline samples in the clamp-type DAC, the single absorption component was observed at ~0.5 mm/s in the SR-based <sup>174</sup>Yb Mössbauer spectrum. This spectrum reveals that  $\beta$ -YbAlB<sub>4</sub> is in a paramagnetic state at 2.45 GPa and 2 K. This feature of the single-absorption component in the spectrum is maintained up to 4.2 GPa and 2 K. These results indicate the absence of a long-range magnetic order in  $\beta$ -YbAlB<sub>4</sub> at 2 K up to 4.2 GPa.



Fig. 1. SR-based  $^{174}\mathrm{Yb}$  Mössbauer spectrum of  $\beta\text{-}$  YbAlB4.

At ambient pressure and 2 K, we observed the two absorption components in the SR-based <sup>174</sup>Yb Mössbauer spectrum, related to slow relaxation time in the valence fluctuations. Accordingly, the sharp single-absorption component observed in these spectra under pressures suggests that the relaxation time in the valence fluctuations is much faster than the energy resolution of the SR-based <sup>174</sup>Yb Mössbauer spectroscopy.

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### Magnetic Phase Diagram of EuPtSi

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EuPtSi crystallizes in a cubic chiral structure ( $P2_13$ ), in which the divalent Eu (J=7/2) ions form a trillium lattice, the same arrangement with the Mn atoms in MnSi. EuPtSi magnetically orders at  $T_N=4.05$  K in a first-order-like transition [1,2], but the magnetic structure is unknown yet.

We study the magnetic phase diagrams of EuPtSi by means of magnetization measurements at low temperatures, and the results are shown in Fig.1. The magnetization M(H) at T=0.25 K increases almost linearly with H and saturates at ~3 T to the value ~7.2 µ<sub>B</sub>/Eu for all directions. When H is applied parallel to [100] or [111], a field-induced phase (A-phase) appears in the intermediate field region at temperatures T>0.25 (H||[100]) and T>0.4 K (H||[111]), characterized by a double-peak structure in dM/dH at  $H_{A1}\sim1$  T and  $H_{A2}\sim1.5-1.8$  T. No such intermediate phase appears for H||[110], and preliminary measurements reveal that the A-phase is absent for H||[112], either.

At present, the low-temperature boundary of the A-phase is still ill defined. On cooling below ~0.8 K, a large hysteresis develops in the critical fields  $H_{A1}$  and  $H_{A2}$  with a concomitant smearing in the dM/dH peak structures. No signature of the A-phase is detected in dM/dH below 0.25 K for both H||[100] and H||[111]. In order to seek the low-temperature boundary of the A-phase, we measured temperature dependences of the magnetization M(T) and the heat capacity C(T) at various magnetic fields. A clear disparity is observed in the zero-field-cooled (ZFC) and field-cooled (FC) data of M(T) in the A-phase region below ~1 K, but no appreciable anomaly is found in the FC data. Meanwhile, new features are found in both M(T) and C(T) near 0.5 K in low fields.

Very recently, a giant additional Hall resistivity has been observed in the A-phase [3], which bears a similarity with the skyrmion phase in MnSi. Whereas the skyrmion of MnSi appears in all directions, the A-phase in EuPtSi is highly anisotropic.



Fig. 1. Phase diagram of EuPtSi for the three principal field directions.

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### Nonreciprocal magnons in noncentrosymmetric magnets

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Lack of inversion symmetry in magnetically ordered systems may result in quite intriguing magnetic excitations, such as nonreciprocal propagation of magnons. The nonreciprocal magnons appears as "shifted" dispersion relation in the magnetic excitation spectrum, and the "shift" direction is given by the symmetry of the system. Recently, in the induced ferromagnetic (fully polarized) state of the chiral magnet MnSi, we observed clear unidirectional shift of the magnon dispersion relation away from the ferromagnetic zone center where the magnetic Bragg reflection is observed [1]. In this talk, we will discuss our new observation of the linear crossing of magnon dispersions at the antiferromagnetic zone center in the noncentrosymmetric antiferromagnet  $\alpha$ -Cu<sub>2</sub>V<sub>2</sub>O<sub>7</sub> [2].

The single crystal neutron inelastic scattering experiment was performed using SPINS and BT-7 triple-axis spectrometers, as well as DCS disc-chopper spectrometer, installed at NIST Center for Neutron Research. The base-temperature inelastic scattering spectrum is shown in Fig. 1(a). The two non-degenerated branches were clearly observed, in contrast to the centrosymmetric antiferromagnet in which the two should degenerate. They are shifted in the opposite direction around the antiferromagnetic zone center Q = (0, 2, 0), where the magnetic Bragg reflection appears, and hence the linear crossing of the two branches is seen at the zone center. The 16-sublattice linear spin-wave calculation reproduces the shifted dispersion relations as shown in Fig. 1(b). It was further found that the external magnetic field changes the spin-wave energies of the two branches in opposite way, as if they have opposite spins. From the observed opposite shift of the magnon branches, we conclude that the magnons in this system would show intriguing magnon-birefringence effect, which may be of technological interest for spintronic applications [3].



Fig. 1. (a) Observed neutron inelastic spectrum in  $\alpha$ -Cu<sub>2</sub>V<sub>2</sub>O<sub>7</sub>. (b) Calculated scattering cross-section for the 16-sublattice model.

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# Macroscopic order parameters for antiferromagnetic phases by cluster multipole moments

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We introduce a theoretical framework which provides a novel concept, cluster multipole (CMP), to characterize macroscopic magnetization of antiferromagnets and identify the order parameters which characterize certain type of antiferromagnetic (AFM) structures inducing the anomalous Hall effect (AHE) [1]. The modern formalism of the intrinsic anomalous Hall conductivity (AHC) provides profound insight into the AHE being closely related to the topology of one-electron energy bands [2-3]. Whereas the AHE is usually observed in ferromagnets and explained as an outcome of the macroscopic dipole magnetization, the AHE has been studied also for certain noncollinear AFM states [4-5]. In particular, a large AHC was recently discovered for the AFM states in  $Mn_3Z$  (Z=Sn, Ge), whose magnetic geometry has no uniform magnetization [6-8]. We applied the CMP theory to the noncollinear AFM states of  $Mn_3Z$  (Z=Sn, Ge) and  $Mn_3Ir$  and show that the AHE is associated with the cluster octupole moments which belong to the same symmetry as the magnetic dipole moments (Fig. 1).

We further provide a new method to generate a complete orthonormal magnetic structure basis set, classified according not only to the symmetry but also to CMP for given crystal structure [9] (Fig. 2). The theory generalizes the multipole theory for macroscopic physical phenomena in magnetic ordered states, such as AHE and electromagnetic effect.



Fig 1. Octupolar moment dependence of the AHE in  $Mn_3Z$ 



Fig 2. Orthonormal magnetic structures classified according to multipole and toroidal moments..

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# Cluster multipole theory for large magneto-optical Kerr effect in Mn<sub>3</sub>Sn

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Since its discovery in 1877 [1], magneto-optical Kerr effect (MOKE) has been widely used as a powerful probe of magnetic properties of materials. Usually, MOKE is observed in magnets having large magnetization, such as ferromagnets or ferrimagnets. However, contrary to this common wisdom, it has been recently shown that Mn<sub>3</sub>Sn with coplanar antiferromagnetic spin structure exhibits very large MOKE [2].

For Mn<sub>3</sub>Sn, it has been shown theoretically [3] and experimentally [4] that the anomalous Hall effect is also extremely large. To understand its origin, we have recently introduced a quantity which we call "cluster multipole", and found that macroscopic "octupolarization" induces the large anomalous effect in this system [5].

In this talk, we will discuss how the octupolarization induces large MOKE in antiferromagnets and weak ferromagnets. While magnetization (dipole) is directly coupled with angular momentum of electrons in ferromagnets, spin-orbit coupling does not directly couple octupolarization with angular momentum. Therefore, the distribution of positive and negative helicity states in the electronic structure is more complicated in antiferromagnets than in ferromagnets, giving rise to characteristic oscillation in the Kerr rotation angle. We also found similar oscillation in the chemical potential dependence of the anomalous Hall conductivity, which is good news for realizing large anomalous Nernst effect in antiferromagnets [6].



Fig. 1. Cluster multipole (octupole in Mn<sub>3</sub>Sn).

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# Magnetism and Superconductivity of Icosahedral Quasicrystals

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Heavy fermion intermetallic compounds have attracted much interest to study unconventional superconductivity and non-Fermi-liquid states, related to quantum criticality. Until very recently, quantum criticality has been intensively studied in only crystalline materials. Quasicrystals possess long-range, quasi-periodic structures with diffraction symmetries forbidden to crystals. Recently, a new type of icosahedral Yb quasicrystal and approximant was discovered [1]. The Au-Al-Yb quasicrystal with Tsai-type cluster exhibits novel quantum critical behavior as observed in Yb-based heavy fermion materials with intermediate Yb valence, while the Au-Al-Yb approximant shows heavy Fermi liquid behavior [2]. Furthermore, quantum critical phenomenon of the Au-Al-Yb quasicrystal is remarkably robust against hydrostatic pressure, related to the critical state unique to the quasicrystal. By contrast, the Au-Al-Yb approximant shows heavy fermion behavior, very sensitive to hydrostatic pressure and quantum criticality of the approximant is induced by pressure [3]. Therefore, the quantum critical state of the Au-Al-Yb quasicrystal might correspond to an electronic state unique to the quasicrystals. Interestingly, quantum criticality of the Au-Al-Yb quasicrystal seems to be closely related to heavy fermion crystalline compound and the icosahedral Yb quasicrystals and approximants shed a new light on strongly correlated electrons in quasicrystals. Studying the magnetism of icosahedral Yb quasicrystals and approximants by substitution of Yb ligands, we have found that the Au-Al-Yb system is located near the border of the valence change [4]. In the process of material research [4-7], we have found superconductivity of Au-Ge-Yb approximants with Tsai-type cluster for the first time [8,9]. Furthermore, we have confirmed the emergence of bulk superconductivity of Al-Zn-Mg quasicrystal at a very low transition temperature of  $T_{\rm c} = 0.05$  K [10]. Comparison between the quasicrystal and the approximants demonstrate that the effective interaction between electrons remains attractive under variation of the atomic arrangement. In superconducting quasicrystal, the fractal geometry might interplay with superconductivity: fractal superconductivity [11].

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# New Quantum Criticality in Yb-Based Quasicrystal and Approximant Crystal

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Unconventional quantum criticality discovered in heavy-electron quasicrystal (QC) Yb<sub>15</sub>Al<sub>34</sub>Au<sub>51</sub> has attracted great interest [1]. The criticality is common to those observed in heavy-electron periodic crystals YbRh<sub>2</sub>Si<sub>2</sub>,  $\beta$ -YbAlB<sub>4</sub>, and  $\alpha$ -YbAl<sub>1-x</sub>Fe<sub>x</sub>B<sub>4</sub> (x=0.014), which is well explained by critical Yb-valence fluctuations (CVF) [2]. The CVF theory based on the minimal model for the QC and the approximant crystal (AC) Yb<sub>14</sub>Al<sub>35</sub>Au<sub>51</sub>, which consist of concentric shell structures with Yb and Au-Al cluster (Tsai-type cluster), has shown (a) robustness of the quantum criticality in the QC under pressure, (b) emergence of the same criticality even in the AC when applied pressure is tuned, and (c) wider quantum critical region in the *T-P* phase diagram of the QC than that of the AC [3,4]. These (a)-(c) have actually been observed by recent experiments [1,5].

To get insight into the mechanism of the emergence of the unconventional quantum criticality observed in the pressurized AC as well as the QC, the AC is analyzed theoretically [6]. By constructing the periodic Anderson model on the AC, which has the periodic arrangement of the bcc structure of the Tsai-type cluster, the heavy quasiparticle band is shown to be formed near the Fermi level because of strong correlation of 4f electrons at Yb. This is the first clarification of the electronic state of the Yb-based AC with an intermediate valence of Yb, since the band-structure calculation has not been performed because of too-many number of atoms per unit cell. We find that charge-transfer mode between 4f electron at Yb on the 3rd shell and 3p electron at Al on the 4th shell in the Tsaitype cluster is considerably enhanced with almost flat momentum dependence. Then, we applied the mode-mode coupling theory of the CVF under magnetic field developed recently [7] to the chargetransfer mode in the AC. The result shows that magnetic as well as valence susceptibility exhibits  $\chi \sim$  $T^{0.5}$  for the zero-field limit and is expressed as a single scaling function of the ratio of temperature to magnetic field T/B over four decades even in the AC when applied pressure is tuned [6]. The T/Bscaling, which is essentially the same as that observed in  $\beta$ -YbAlB<sub>4</sub> [8] and  $\alpha$ -YbAl<sub>1-x</sub>Fe<sub>x</sub>B<sub>4</sub> (x=0.014) [9], has also been detected recently in the pressurized AC [5] and the QC [10]. The key origin is clarified to be due to strong locality of the CVF and small Brillouin zone reflecting the large unit cell, giving rise to the extremely-small characteristic energy scale of the CVF. This also gives a natural explanation for the quantum criticality in the QC [1] corresponding to the infinite limit of the unitcell size of the AC. Systematic substitutions of Al and Au to Ga and Cu respectively in the QC and AC have recently revealed that Yb<sub>15</sub>Al<sub>34</sub>Au<sub>51</sub> is indeed located at just the point where the Yb valence starts to change sharply as a function of the lattice constants [10]. This feature is also explained by the theory of the CVF, which supports that this material is located at the valence QCP [11].

In the presentation, we discuss these newly-clarified aspects of quantum critical phenomena and the key origin of the unconventional criticality commonly observed in Yb-based periodic and aperiodic systems [12]. This presentation is based on the work done in collaboration with K. Miyake.

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# Probing occupied 4f-orbital symmetry by linear dichroism in angle-resolved core-level photoemission

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We have discovered that the atomic-like multiplet-split structure in *angle-resolved* core-level photoemission spectra shows linear dichroism (LD) reflecting the anisotropic outer 4f spatial distributions determined by the crystalline-electric-field (CEF) splitting for tetragonal [1] and cubic [2] Yb compounds. LD in the angle-resolved core-level photoemission spectra should not be restricted to the Yb compounds but widely be seen in many rare-earth elements since it is due to the anisotropy in the Coulomb and exchange interactions between the electrons in the single site. Actually, we have successfully observed the LD in Sm<sup>3+</sup> 3d<sub>5/2</sub> core-level spectra along the [001] direction for tetragonal SmCu<sub>2</sub>Si<sub>2</sub> [3], of which the CEF-split ground

state is found to be in the  $\Gamma_7$  symmetry.

Furthermore, LDs have successfully been detected for cubic Pr compounds. We have performed the polarization-dependent angleresolved hard x-ray core-level photoemission at BL19LXU of SPring-8 with hv = 7.9 keV, which is high enough for probing the bulk electronic structure overcoming the surface sensitivity of The experimentally lowest photoemission spectroscopy [5]. temperature was ~5 K. Figure 1 shows the polarization-dependent Pr 3d<sub>5/2</sub> photoemission spectra of PrIr<sub>2</sub>Zn<sub>20</sub> along the [100] direction. Although the LD is subtle but finite, spectral simulations on the basis of a single-site  $Pr^{3+}$  ion model assuming the  $\Gamma_3$  ground state can reproduce our experimental results [4]. The sign of LD is flipped for  $PrB_6$  with the  $\Gamma_5$  ground state, which suggests the power of the polarization-dependent angle-resolved core-level photoemission for probing occupied local 4f-orbital symmetry. Recently, we have also observed a clear LD in line with the  $\Gamma_1$  ground state for PrBe<sub>13</sub>.

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with simulations [4].

# AGround-state phase diagram in the Kugel-Khomskii model with finite spinorbit interactions

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Strongly correlated electron systems with orbital degrees of freedom have attracted much interest. One of the intriguing examples is the series of the honeycomb-layered compounds,  $A_2IrO_3$  (A=Li, Na) with 5d electrons [1] and  $\alpha$ -RuCl<sub>3</sub> with 4d electrons [2]. In these compounds, a strong spinorbit interaction should cause the anisotropy in the exchange coupling between spins. Then, the compounds are regarded as the candidates for realizing the Kitaev model [3]. Recently, fermionic response has been discussed in  $\alpha$ -RuCl<sub>3</sub> at finite temperatures [4], which stimulates further theoretical and experimental investigations on the Kitaev and related models [5]. A simple question that naturally arises is whether or not the finite spin-orbit interaction yields interesting ground-state and low temperature properties characteristic of the Kitaev model.

To clarify this, we consider the spin-orbital model with the Kugel-Khomskii type superexchange interaction on the honeycomb lattice. This model should be derived from the multi-orbital Hubbard model with  $t_{2g}$  orbitals in the strong coupling limit. Using the cluster mean-field approximations with six- and ten-site sites [6], we treat the superexchange interaction and spin-orbit coupling on equal footing. Then, we clarify how some magnetically ordered states compete with each other. Furthermore, we discuss the stability of the spin liquid state realized in a strong spin-orbit coupling limit (Kitaev limit).

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# Ordering and Fermi-Liquid Properties due to Orbital-Dependent Hybridization in Sm Compounds

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It has long been known that the compounds including Sm ions often display puzzling electronic properties owing to c-f hybridization at low temperatures. In this presentation, we shall summarize our activities supported by the "J-Physics" project, concerning the mechanisms of two typical phenomena in Sm compounds.

The first one is an ordering phenomenon in SmRu<sub>4</sub>P<sub>12</sub>. It has been believed that an anomalous magnetic field dependence of the antiferromagnetic transition is a consequence of forming a novel spin-charge composite order phase in magnetic fields. Making use of a Kondo-Lattice model derived from the p-f hybridization for the RRu<sub>4</sub>P<sub>12</sub> system, we extended the analysis to the transport properties, considering the scattering of conduction electrons by the order-parameter fluctuations [1]. Then, it was shown that complicated temperature and field dependences of the resistivity observed in SmRu<sub>4</sub>P<sub>12</sub> are caused by partial gap suppression due to the mixture of spin and charge order parameters. In addition, we predicted that the spin-dependent quasiparticle state formed in the composite phase results in a significant enhancement of the spin conductivity in magnetic fields.

Quite recently, we are studying on a possible carrier-doping effect in the p-f derived Kondo lattice model at zero magnetic field. It is found that the normal antiferromagnetic phase is unstable to hole doping and easily replaced by the composite phase. The mechanism of emerging the composite phase at zero field will be discussed in connection with site dependences of spin and charge order parameters.

The second topic is the field-insensitive heavy fermion (HF) in some Sm compounds such as  $SmOs_4Sb_{12}$ ,  $SmTa_2Al_{20}$  and so on. Considering that  $Sm^{2+}$  has the f<sup>6</sup> spin-orbit coupled ground singlet, we introduced a two-orbital impurity Anderson model with an effective f<sup>2</sup> singlet ground state in the localized limit. Then we studied the ground-state and Fermi-liquid properties of the model by employing the numerical renormalization method [2]. It was shown that a quantum critical point (QCP) between the local- and Kondo-singlet phases always takes place as a function of the hybridization in the model. In the vicinity of the QCP, a HF state with a vanishingly small Wilson ratio was found to be realized and to possess an intermediate average occupancy of f electrons (Fig. 1). The relevance of those characteristics of the effective model with the real Sm compounds will be discussed.



Fig. 1.  $\gamma = C / T$  and the Wilson ration W of the effective Anderson model.

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# Methods of Sparse Modeling in Quantum Many-Body Physics

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Sparse modeling refers to a concept and methodology for finding a few number of relevant parameters that well explains a given data set. This data-science approach enables us to extract useful information even from low quality data that include measurement errors and noise. Its power has already been established by successful applications in various fields, such as MRI and astronomical observations.

Recently, we have applied sparse modeling to quantum many-body physics. Quantum Monte Carlo (QMC) methods are important tools of wide applicability in studies of quantum many-body systems. In practical computations, however, one frequently faces two problems: (1) ill-conditioned analytical continuation of imaginary-time data to real-frequency spectra and (2) massive high-dimensional data for multiple-time correlation functions. The former reduces reliability of even highly accurate QMC data regarding dynamical properties, and the latter limits applications of sophisticated QMC simulations to realistic material calculations.

I first present a new method of analytical continuation based on sparse modeling, and show that a stable analytical continuation becomes possible [1]. This method could alter the de facto standard tool, namely, maximum entropy method. A detailed analysis of analytical continuation reveals the existence of a quite efficient basis set which can express the single-particle and two-particle Green function in an extremely compact form [2,3]. It suggests that the new basis could improve efficiency of QMC measurements and diagrammatic calculations including vertex parts. The sparse modeling approach to quantum many-body physics will be expanded further [4-6].

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# Spin Nematic Phases in Low-Dimensional Quantum Antiferromagnets

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The spin nematic phase, which is a kind of multipole phases, has attracted a lot of interest in the field of the strongly correlated electron systems, as well as the quantum spin liquid phase. Using the numerical exact diagonalization, the density matrix renormalization group (DMRG) calculation, and the finite-size scaling analysis, it is found that some spin nematic and spin liquid phases appear in the anisotropic and/or frustrated quantum spin systems.

In our previous work, quasi-one-dimensional quantum spin systems with the easy-axis anisotropy in magnetic field are theoretically investigated using the numerical exact diagonalization, the density matrix renormalization group (DMRG) and the finite-size scaling analysis[1]. It was found that a field-induced nematic phase appears at some critical field in the anisotropic spin ladder and the mixed spin chain. The nematic phase is characterized by the power-law decay in the correlation function of the second-order spin moment. In addition at some higher critical field a quantum phase transition can occur to the conventional field-induced Tomonaga-Luttinger liquid. Several typical magnetization curves calculated by DMRG are presented. We will also propose an experiment to observe these field-induced transitions by NMR measurements. The field-induced incommensurate order observed in azurite, which is modeled by the distorted diamond chain, by the recent NMR measurement will be also discussed.

Recently the field-induced nematic phase was observed on the frustrated spin ladder system[2,3]. So we study on a frustrated spin ladder system[4], using the numerical diagonalization and DMRG. As a result, it is found that several exotic quantum phases, including the spin-nematic liquid phase. We also report some exact eigenstates of the present model and present several interesting phase diagrams[5].

We also obtained some new results about the magnetization process of frustrated systems; the triangular lattice quantum antiferromagnet with the next-nearest-neighbor interaction[6], or with the lattice distortion[7,8]. Those results will be presented.

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# Phonon Hall effect by extended cluster multipoles

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A heat current perpendicularly flowing to a magnetic field induces a transverse temperature gradient in the third perpendicular direction. This is the thermal Hall effect (THE). It is trivial in a metal because of Lorentz force, whereas it is not so obvious in an insulator, since there are no mobile charges such as electrons nor holes to assist the thermal transport. Nevertheless, it is reported that a non-magnetic insulator, Ba<sub>3</sub>CuSb<sub>2</sub>O<sub>9</sub> (BCSO), shows the THE [1]. Strictly speaking, the BCSO is magnetic and is in the quantum spin liquid state. Its hexagonal perovskite-type structure contains a honeycomb network of Cu<sup>2+</sup> ions. The hexagonal symmetry is kept down to temperatures much lower than the magnitude of magnetic exchange interaction (~50 K). The BCSO does not show magnetic long-ranged order and has the spin gap of about 50 K in energy [2]. Hence, the heat transport in temperatures lower than 50 K must be dominated by phonons instead of spins. So that the THE in the BCSO is the "phonon Hall effect" (PHE). Here the question arises: What is the origin of the PHE in the BCSO?

The PHE is originally observed in a non-magnetic insulator, terbium-gallium-garnet (TGG) [3,4]. This phenomenon originates from a resonant scattering of phonons by  $Tb^{3+}$  ions with total angular momentum J=6 [5]. The crystal electric field (CEF) is expanded with respect to lattice strains. Thanks to the large J, a modulation of the CEF is a function of J and lattice strains. This is the origin of spin-phonon coupling in the TGG. On the other hand, however, the origin of THE in BCSO is still uncovered, since the BCSO does not contains any ion with large J and is composed of only spin 1/2, which is isotropic in the charge degree of freedom.

In this study, we will propose that an "orphan spin" is the key of the PHE in the BCSO. The orphan spin is an additional  $Cu^{2+}$  spin located in the center of  $Cu^{2+}$  hexagonal. It is said that there are about 5%–16% of the  $Cu^{2+}$  orphan spins in BCSO, and the thermal conductivity in low temperatures is dominated by the orphan spins [1]. We find that an elongation of  $Cu^{2+}$  hexagonal with an orphan spin, which may correspond to the Jahn-Teller distortion in a non-stoichiometric BCSO, leads to a charge redistribution with a quadrupole symmetry. The quadrupole electric field can couple to lattice strains. This must be the origin of spin-phonon coupling in the BCSO. Since the  $Cu^{2+}$  hexagonal is a spin-1/2 cluster, a spin-flip scattering of phonon is also possible. In short, the extended cluster multipole of  $Cu^{2+}$  hexagonal with an orphan spin must be the origin of PHE in the BCSO.

This work is done in collaboration with Prof. H. Kusunose.

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# Current-induced bulk magnetization in elemental tellurium

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The magnetoelectric effect in bulk matter is of growing interest both fundamentally and technologically. Since the beginning of the century, the magnetoelectric effect has been studied intensively in magnetic materials that break time-reversal symmetry. However, magnetoelectric phenomena in materials that preserve time-reversal symmetry without any magnetic order remain almost unexplored.

Here we show the observation of a new class of bulk magnetoelectric effect, by revisiting elemental trigonal tellurium, which is a nonmagnetic semiconductor without inversion symmetry. We demonstrate that using nuclear magnetic resonance (NMR) techniques, elemental tellurium exhibits current-induced magnetization [1]. Figure 1 shows the appliedcurrent dependence of the <sup>125</sup>Te-NMR spectrum of elemental tellurium. The position of the <sup>125</sup>Te-NMR spectrum shifts in proportion to the applied current density, as shown in this figure. This clearly indicates that the applied current induces bulk magnetization in elemental tellurium.

We overview the general origin of the current-induced magnetization effect in materials with time-reversal symmetry. We also discuss the role played by spin-orbit interaction in the current-induced magnetization effect in elemental tellurium, showing <sup>125</sup>Te-NMR data under pulsed current at a pressure of 20 kbar.

This work has been in collaboration with Yuri Shimokawa (Tokyo University of Science), Tetsuya Furukawa (Tokyo University of Science), and Kaya Kobayashi (Okayama University).

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# **Tests for Magnetoelectric Effects on Antiferromagnetic Metals**

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The research conducted by Group C01 is focused on "spatially-extended multipoles", which are defined as a unit of spins or charges arranged on a certain number of ions; *i.e.*, spin or charge clusters which can be described as "extended/augmented magnetic or electric multipoles". For example, an all-in and all-out spin states on octahedrons in pyrochlore oxide compounds can be regarded as extended magnetic octupoles (even parity) with opposite directions. Vertex like spin order on a hexagon in a honeycomb lattice can be described as an odd-parity extended magnetic multipole called a toroidal moment [1]. Group C01 studies exotic cross correlations and dynamic responses among electricity, magnetism, and elasticity expected in the ordered states of such extended multipoles, particularly occurring in metallic systems.

We present here a progress report on our studies of experimental tests for a new magnetoelectric (ME) effect of "electric-current-induced magnetization" expected in certain antiferromagnetic (AF) metals. This phenomenon was theoretically predicted to occur when the global inversion symmetry of a system is spontaneously broken by the AF ordering, where the ordered state can be described as possessing a uniform component of a toroidal moment t [1, 2]. We have thus far investigated three AF metals, UNi<sub>4</sub>B (Space group: *Cmcm*, No. 63,  $D_{2h}^{17}$ ;  $T_N = 20.4$  K) [3,4], CeRh<sub>2</sub>Si<sub>2</sub> (*I4/mmm*, No. 139,  $D_{4h}^{17}$ ;  $T_{N1} = 35$  K,  $T_{N2} = 24$  K) [5], and CeRu<sub>2</sub>Al<sub>10</sub> (*Cmcm*, No. 63,  $D_{2h}^{17}$ ;  $T_N = 27$  K) [6, 7], and found that all of them show such a ME effect in their AF ordered states. Common characteristics among observed behaviors are (i) the current-induced magnetization  $\Delta M$  is perpendicular to the direction of electric currents I, (ii)  $\Delta M$  is directly proportional to I, and (iii) the rate of induction of  $\Delta M$  is on the order of ~ 10<sup>-7</sup> - 10<sup>-9</sup>  $\mu_B/(U \text{ or Ce})$  per unit current density (in units of kA/m<sup>2</sup>). The obtained experimental results have confirmed that a uniform magnetization component can be induced by applying electric currents in certain types of AF metals, and its magnitude and direction are closely coupled with the magnetic structure. This feature is considered to be essentially consistent with the theoretical prediction [2]. However, we also observed some inconsistencies that  $\Delta M$  is induced in an unexpected geometry between I and t in UNi<sub>4</sub>B [8], and moreover, in CeRh<sub>2</sub>Si<sub>2</sub> and CeRu<sub>2</sub>Al<sub>10</sub>  $\Delta M$  occurs in the AF states where the global space-inversion symmetry is conserved if the known magnetic structures are correct. We will discuss the implications of these results and future research direction.

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# B-NMR study on paramagnetic and toroidal magnetic ordered states in UNi<sub>4</sub>B

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UNi<sub>4</sub>B exhibits an antiferromagnetic (AFM) ordering at  $T_N = 20$  K and shows highly anomalous magnetic behaviour in the AFM state[1,2]. It was reported that the crystal structure of this material is the hexagonal CeCo<sub>4</sub>B-type crystal structure described by the space group P6/mmm[1]. From the neutron scattering measurement, the uranium magnetic moment forms a vortex-like structure with multiple propagation vector Q in the [0001]-plane, so called a ferro-toroidal ordering [3]. The magnetic structure is interpreted that two-third uranium moments form 60° rotated spin structure, whereas one-third uranium moments remain to be a paramagnetic state[3]. The paramagnetic moments order below 330 mK [4].

Recently, Hayami et al. pointed out theoretically that the broken local-inversion symmetry at magnetic-ion sites possibly causes a toroidal ordering. They predicted that exotic magnetotransport and magnetoelectric effects might occur under the toroidal order, such as UNi<sub>4</sub>B. However, Haga and co-workers reported that the crystal structure is not the hexagonal (Space group; P6/mmm) but the orthorhombic (Space group; Cmcm)[5]. It is unclear whether or not the difference of the crystal structure affects the toroidal magnetic structure.

Here we report that B-NMR results under magnetic fields of 5 T and 1 T. At the low magnetic field of 1T, <sup>11</sup>B-NMR line cannot be explained by hexagonal structure, but rather by orthorhombic structure with either Cmcm [5,6] or Cmc2<sub>1</sub> [6] space group. At the high magnetic field of 5T, on the other hand, the NMR spectrum shows magnetic dipolar spectral broadening and cannot be distinguishable between hexagonal structure and orthorhombic one. In addition, our preliminary NMR data do not contradict with the magnetic structure reported previously. These results suggest that the deformation from the hexagonal structure to orthorhombic structure does not have a potent influence in magnetic properties.

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# Resonant x-ray scattering study on hybridized orbital states in *d*- and *f*- electron system

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Hybridized orbital states between the metal and the ligand ions play a crucial role in the physical properties in the *d*- and *f*- electron system. In fact, the orbital hybridization between the conduction electron and the *f*-electron, so called *c*-*f* hybridization, becomes a key parameter in the rare earth compounds. Spatially extended multipole, which is noted in this research field, is also composed of the orbital hybridization. Hence the study of hybridized orbital state is an important issue. Resonant X-ray scattering (RXS) technique is a powerful tool for observing the multipole orderings in the *d*- and *f*- electron system [1]. The resonant signal at particular absorption energy can reveal element- and orbital-selective information of electronic state of the absorbed ion. Here, the RXS studies on the manganite superlattice and the skutterudite PrRu<sub>4</sub>P<sub>12</sub> will be presented.

Manganite superlattice  $(LaMnO_3)_m(SrMnO_3)_n$  (LmSn) has widely been investigated, and it is noted that the physical properties of the LmSn superlattice strongly depend on the stacking structure of the LaMnO<sub>3</sub> and SrMnO<sub>3</sub> layers [2]. Then a large negative magnetoresistance, which is realized for the first time by the fabrication of the superlattice structure, was newly discovered [3]. Here we noted the L2S2 superlattices with different physical properties, and the charge disproportionation of Mn 3d and O 2p has been investigated by the RXS technique. It elucidated that the charge ordering is changed from the charge disproportionation of Mn 3d to that of O 2p in these manganite oxides. It indicates that the strong p-d hybridization on the O 2p charge disproportionation is related to the conductivity in the superlattice system. The oxygen magnetism reflecting the charge disproportionation of O 2p was also clarified.

Filled skutterudite PrRu<sub>4</sub>P<sub>12</sub> exhibits a metal-insulator (MI) transition at  $T_{MI} = 63$  K [4], and has attracted much attention to an origin of the transition, i.e. charge density wave [5], antiferro-hexadecapole order [6], and so on. Neutron scattering study elucidated the presence of strong orbital hybridization between Pr 4f and P 3p (p-f hybridization), which causes the formation of staggered f-electron order below  $T_{MI}$  [7]. Hence the p-f hybridization is expected to be essential for the MI transition. In order to clarify the hybridized orbital state in the unconventional ordered phase, resonant x-ray scattering has been performed at the Pr L<sub>3</sub>-edge (2p -> 5d transition) and at the P K-edge (1s -> 3p transition). Resonating energy spectra were found at the Pr L<sub>3</sub>-edge and at the Pr L<sub>3</sub>-edge. This result is expected to be related with the strong p-f hybridization effect on the staggered order phase.

These works have been performed in collaboration with C. Tabata, Y. Yamasaki, H. Yamada, M. Kawasaki, K. Iwasa, and Y. Murakami.

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# Homo-chiral crystallization in inorganic chiral magnetic materials

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

First, our unique crystallization technique for water-soluble inorganic chiral compounds will be presented. By adapting our spontaneous crystallization technique with stirring in combination with careful examination of crystallographic chirality using X-ray, we succeeded in obtaining the centimeter-sized enantiopure single crystals of chiral helimagnetic CsCuCl<sub>3</sub> [3]. Second, our recent progress of homo-chiral crystal growth will be presented. We are developing homo-chiral crystal growth techniques for non-water-soluble chiral magnetic compounds. We will introduce some of the techniques.

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# **Cross-correlated coupling based on augmented multipoles**

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A property of linear responses is governed by underlying symmetry of solids. For example, the magnetoelectric (ME) response requires breaking of both time and inversion symmetries [1], while the magneto-current (Mj) or gyrotropic magnetic (Edelstein) effect requires only inversion symmetry breaking (precisely speaking, it requires gyrotropic point group) with finite electric current [2,3]. Such a symmetry breaking and associated cross-correlated coupling are caused by a built-in crystal structure or a spontaneous symmetry breaking due to an ordering of an electronic degrees of freedom.

Concerning the latter, arbitrary type of electronic degrees of freedom can be expressed by four fundamental multipoles, *i.e.*, electric (E), magnetic (M), electric toroidal (ET), and magnetic toroidal (MT) multipoles [4]. We extend a concept of conventional multipoles [5] in two ways: (i) cluster multipole and (ii) hybrid multipoles [6], both of which are categorized as "augmented" multipoles. The former is an alternative view of characteristic alignment of magnetic dipoles, *i.e.*, plural magnetic dipoles over different sites (a cluster) are regarded as single multipole object [7,8,9]. For instance, a vortex-like magnetic structure can be characterized by the MT dipole. On the other hand, the latter is defined over different atomic orbitals, e.g. *p-d* hybridized orbitals. It should be noted that the cluster multipole, e.g. a MT dipole, always accompanies a M dipole order by definition, while the hybrid multipole is a pure quantum object without having any M dipoles. Thus, the latter is usually hidden in condensed matter.

I will demonstrate some examples of the interplay between "augmented" multipoles and crosscorrelated couplings [6,7,8,10,11]. These works are in collaboration with Y. Yanagi, S. Hayami, and Y. Motome.

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# Surface Properties of a chiral d-wave superconductor with hexagonal symmetry

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We discuss surface properties in a chiral d-wave superconductor with hexagonal symmetry, one-body Hamiltonian of which possesses the intrinsic spin-orbit coupling identical to the one crucial to the topological structure of the Kane-Mele honeycomb insulator. In the normal state, the spin-orbit coupling causes spontaneous surface spin currents, while in the superconducting state there exist besides the spin currents also charge surface currents, due to the chiral pairing symmetry with broken time-reversal symmetry. Interestingly, the combination of these two currents results in a surface spin polarization, whose spatial dependence is markedly different on the zigzag and armchair surfaces. There are various potential candidate materials, such as SrPtAs, which may exhibit these surface properties.

# **Exploration of Materials with Odd-Parity Multipole Order**

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Kondo effect is one of the most fascinating phenomena in condensed matters. The phenomenon is realized when conduction electrons couple with a localized spin, which is an even-parity magnetic dipole moment. A natural extension of this phenomenon is a quantum transport in a system, in which conduction electrons couple with a higher order electric/magnetic multipole. Among higher order electric/magnetic multipoles, we are particularly interested in odd-parity multipoles such as electric octupole and magnetic quadrupole. This is because one can expect a novel quantum transport induced by the breakdown of the global inversion symmetry in a metal with the odd-parity multipole order. However, there are few examples of actual materials with the odd-parity multiple order, preventing us from detailed studies on exotic quantum transport properties.

Up to now, we have found several novel materials, in which odd-parity multipoles are ordered in a ferroic manner. Examples include  $Pb_2Re_2O_7$  and  $Pb_2Ir_2O_7$  with the electric octupole order [1, 2]. In this presentation, I will introduce our recent research, which identifies a ferroic order of magnetic quadrupole/hexadecapole in BaMn<sub>2</sub>As<sub>2</sub>. Particularly, I will discuss the domain size of the magnetic quadrupole/hexadecapole order. I will also talk about a ferroic order of electric dipole in BaFe<sub>2</sub>Se<sub>3</sub>, which is stabilized by block-type lattice distortions.

This work was done in collaboration with M. Matsubara, K. Emi, S. Imaizumi, T. Aoyama, K. Igarashi, S. Kimura, Y. Hirata, M. Nakajima, and T. Suemoto.

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# Magnetic Properties of YbNi<sub>3</sub>Al<sub>9</sub> and Related Materials with a Non-centrosymmetric Crystal Structure

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Non-centrosymmetric materials have attracted much attention because they show peculiar physical properties caused by asymmetric spin-orbit interaction, such as magnetoelectric effect, chiral magnetism, and parity-mixed superconductivity. For magnetoelectric effect, most of the work has been done on an insulator. Recently, a new current-induced magnetoelectric effect was reported for a metallic UNi<sub>4</sub>B, in which inversion symmetry is broken locally at magnetic Uranium-ion site [1].

To study the current-induced magnetoelectric effect caused by the asymmetric spin-orbit interaction, we have synthesized single crystals of a rare-earth intermetallic compound YbNi<sub>3</sub>Al<sub>9</sub> and related materials by the flux method. YbNi<sub>3</sub>Al<sub>9</sub> crystalizes in chiral crystal structure of trigonal ErNi<sub>3</sub>Al<sub>9</sub>-type with a space group of R32 [2,3]. The crystal symmetry has no inversion center and no mirror plane. The magnetic rare-earth ion forms a honeycomb layered structure, and inversion symmetry is broken at each rare-earth ion site.

In this work, we investigated current-induced magnetization for YbNi<sub>3</sub>Al<sub>9</sub>, ErNi<sub>3</sub>Ga<sub>9</sub> and LuNi<sub>3</sub>Al<sub>9</sub>. We also determined magnetic field-temperature phase diagram using specific heat and magnetization measurements for YbNi<sub>3</sub>Al<sub>9</sub>, ErNi<sub>3</sub>Ga<sub>9</sub> and DyNi<sub>3</sub>Ga<sub>9</sub> [4-6], and measured magnetic structure by neutron diffraction for ErNi<sub>3</sub>Ga<sub>9</sub> and DyNi<sub>3</sub>Ga<sub>9</sub> [5, 6]. The specific heat and the magnetization measurements were performed using the commercial measurement systems of PPMS and MPMS (Quantum Design), respectively. The neutron diffraction measurement was carried out using BL18 SENJU at the MLF in J-PARC.

Here, we report on a magnetic structure and a current-induced magnetization for ErNi<sub>3</sub>Ga<sub>9</sub>. We have found that ErNi<sub>3</sub>Ga<sub>9</sub> is an *Ising*-like antiferromagnet below 6.4 K with a magnetic propagation vector q = (0, 0, 0.5). The magnetic easy axis is parallel to the [0001] direction. The honeycomb structure of the Erbium ion is divided into two triangular magnetic sub-lattices with their moments antiparallel. Interestingly, the electrical current parallel to the [0001] direction induces a magnetization along the [2110] below the magnetic ordering temperature of 6.4 K. The value of current-induced magnetization for the current density reaches about 2 x 10<sup>-9</sup> µ<sub>B</sub>m<sup>2</sup>/A for Er-ion at 2 K. This value is much larger than that reported for UNi4B.

We will address the magnetic properties of YbNi<sub>3</sub>Al<sub>9</sub> and related materials and discuss the observed current-induced magnetization in *Ising*-honeycomb magnet ErNi<sub>3</sub>Ga<sub>9</sub>.

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# Uniaxial Pressure Effect on the Antiferromagnetic Order in the Kondo Semiconductor with Zigzag Chains

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The Ce-based Kondo semiconductors  $CeT_2Al_{10}$  with T = Ru and Os exhibit an antiferromagnetic (AFM) transition at unexpectedly high temperature  $T_N \cong 28$  K, whose origin remains elusive [1]. Hanzawa proposed that it is related with the lack of inversion symmetry along the *b* axis at the Ce site, which allows on-site hybridization of the 4*f* state with 5*d* state [2]. The AFM order is preceded by the opening of a charge-density-wave like gap in the optical conductivity along the *b* axis [3]. Systematic studies of the effects of dilution and electron/hole doping on the magnetic and transport properties, electron tunneling, and spin-gap formation indicated that the hybridization gap is indispensable for the unusual AFM order [4-6].

Recently, we have revealed anisotropic dependence of the AFM transition on uniaxial pressure [7-8]. It is found that  $T_N$  depends linearly on the *b*-axis parameter under uniaxial pressure P//b and hydrostatic pressure (Fig. 1). Furthermore, in the alloy system Ce(Ru<sub>1-x</sub>Os<sub>x</sub>)<sub>2</sub>Al<sub>10</sub> for  $0 \le x \le 1$ , application of P//b largely increases  $T_N$  without changing the Kondo temperature  $T_K$ , as shown in Fig. 2 [8]. This relation in support of the model that a kind of CDW developing along the *b* axis far above  $T_N$  induces the AFM order in these Kondo semiconductors.



**Fig. 1.** Variations of  $T_N$  for CeT<sub>2</sub>Al<sub>10</sub> (T = Ru, Os) under uniaxial and hydrostatic pressures as a function of the orthorhombic lattice parameters *a*, *b*, and *c* [7].



**Fig. 2.** Relation between  $T_N$  and  $T_{\chi m}$  for Ce(Ru<sub>1-x</sub>Os<sub>x</sub>)<sub>2</sub>Al<sub>10</sub> under uniaxial pressures  $P \parallel a, P \parallel b$ , and  $P \parallel c$  [8]. Here,  $T_{\chi m}$  is the temperature at the maximum of magnetic susceptibility  $\chi(T)$  for B//a and B//c, being a measure of Kondo temperature  $T_K$ .

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# Development of *ab-initio* method based on dynamical mean-field approximation and applications

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In this talk, we review our recent achievements on the development of ab-initio method based on dynamical mean-field theory (DMFT) and applications to real materials.

In recent years, transition metal oxides have been studied extensively. An *ab-initio* method based on DFT and DMFT (DFT+DMFT method) has proven its power to describe multi-orbital effects and magnetism in these compounds. In DFT+DMFT calculations, the original model is mapped to an effective quantum impurity problem, which is solved with solved modern numerical techniques such as continuous-time quantum Monte Carlo (CT-QMC) methods. However, only few open-source implementations are available in our community.

We have developed an open-source implementation of CT-QMC methods for solving quantum impurity models with general two-body interactions and spin-orbit coupling [1]. Using the new implementation, we study multi-orbital physics in the heavy *d*-electron compound  $\text{LiV}_2\text{O}_4$  and the 5*d* pyrochlore oxide  $\text{Cd}_2\text{Os}_2\text{O}_7$ . For the sake of experimentalists, we discuss the usefulness and limitation of DFT+DMFT calculations with the modern impurity solver in material research.

In the latter half of the talk, we briefly introduce our "sparse modeling approach" to go beyond the current state-of-the-art *ab-initio* studies on correlated electron systems. The new technology enables to a stable estimation of the spectral function from QMC data and compression of QMC data, which will open a route to the application of modern many-body theories to realistic strongly correlated electron systems.

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## Multipole Physics in Non-Fermi-Liquid State Due to a Crystal-Field Multiplet

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Crystal-field multiplets coupled to conduction electrons have been considered as a key to understanding anomalous metallic behavior observed in various heavy-fermion systems and investigated as an important problem of multipole physics for the last two decades. In particular, it is expected that the two-channel Kondo effect can be realized in U or Pr-based compounds, which is associated with the quadrupole degrees of freedom of a non-Kramers doublet ground state in the  $f^2$  configuration [1, 2].

At this meeting, we report a recent theoretical study of a crystal-field triplet as another candidate of the multichannel (multipolar) Kondo effect, taking account of the  $O_h \Gamma_5$  triplet as a Pr ground state in a cubic crystal field environment. The low-temperature physics is described by an S = 1 local spin coupled to conduction electrons with fourfold degenerate orbital components of a total angular momentum j = 3/2 that correspond to partial waves with the  $\Gamma_8$  representation. In real systems, an effective exchange interaction between the j = 3/2 electrons and the S = 1 spin consists of magnetic  $j \cdot S$  and quadrupolar  $q \cdot Q$  types. The latter is a scalar product of five components of quadrupole tensors. The earlier studies showed that both magnetic and quadrupolar exchange couplings become relevant at low temperatures, leading to a power law divergence of the temperature dependent magnetic impurity susceptibility and specific heat coefficient [3, 4]. This is different from the non-Fermi-liquid behavior with the logarithmic temperature dependence owing to the two-channel (quadrupolar) Kondo effect.

At the next step of this study, we also examine a quadrupole moment induced by an external field as a peculiarity of the S = 1 spin. We elucidate how the j = 3/2 Kondo effect brings about a nontrivial temperature dependence of the field-induced quadrupole and the non-Fermi-liquid behavior mentioned above is closely related to both magnetic and quadrupolar correlations. In the absence of the inversion symmetry, the induced quadrupole is coupled to an electric dipole with the same  $T_d$  (or T) point-group irreducible representation that is dependent on the field direction [5]. This indicates another possibility of the Kondo effect correlated with a local electric field.

Recently, triplet ground states have been identified in the concentrated Pr  $(4f^2)$  compounds PrCu<sub>4</sub>Ag [6] and PrCu<sub>4</sub>Au [7] (heavy-fermion properties were reported for the latter [8]). For future experiments, dilution of Pr by La is strongly urged to clarify the Pr single-ion effect and confirm the relevance of the quadrupole degrees of freedom to the correlations of f-electrons with conduction electrons.

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# **Exploration of Novel Superconductors with 5d Transition Metals**

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We will give an overview of novel superconducting materials with the emphasis on local spatial symmetry and strong spin-orbit coupling, which play an important role to produce exotic superconducting state.

 $Mg_2Ir_3Si$  --- We discovered a novel Ir-based superconductor  $Mg_2Ir_3Si$  with  $T_c = 7$  K. This novel compound crystallizes in the hexagonal  $Mg_2Cu_3Si$ -type structure (space group  $P6_3/mmc$ ,  $D_{6h}^4$ , #194), which is the fully ordered C14 Laves phase. The structure consists of the breathing Kagome network of Ir. Thus, the spatial inversion symmetry is locally broken at the Ir site.

*IrIn*<sub>2</sub> --- We found that IrIn<sub>2</sub> exhibit superconductivity at  $T_c = 2.2$  K. IrIn<sub>2</sub> crystallizes in the orthorhombic Mg<sub>2</sub>Cu-type structure (space group *Fddd*,  $D_{2h}^{24}$ , #70). The structure consists of infinite Ir zigzag chains. The spatial inversion symmetry at the Ir site is locally broken because of the IrIn<sub>8</sub> square anti-prism coordination.

 $Mo_6Se_8$  --- Chevrel phase compounds  $Mo_6S_8$ ,  $Mo_6Se_8$ , and  $Mo_6Te_8$  have been synthesized via low-temperature topotactic reactions.  $Mo_6S_8$  and  $Mo_6Se_8$  exhibited superconductivity at  $T_c = 1.8$ and 6.8 K, respectively, while superconductivity was not observed in  $Mo_6Te_8$ . Low-temperature specific heat revealed that a large value of electron density of states at the Fermi level as well as the presence of soft phonons, are the key factors in the higher  $T_c$  in  $Mo_6Se_8$ . The structure consists of  $Mo_6$  octahedra, and the spatial inversion symmetry is locally broken at the Mo site.

 $KBi_2 \rightarrow C15$  Laves phase compound KBi<sub>2</sub> crystallizes in the cubic MgCu<sub>2</sub>-type structure (space group  $Fd\overline{3}m$ ,  $O_h^7$ , #227) with the Pyrochlore network of Bi. KBi<sub>2</sub> exhibits superconductivity at  $T_c = 3.6$  K. We found the occurrence of a peculiar reentrant superconductivity that appears when a specific heat treatment was applied. Long-term duration (~500 hours) at room temperature resulted in the disappearance of the reentrant behavior and superconductivity at 3.6 K was restored.

These studies have been performed in collaboration with Kazutaka Kudo, Hikaru Hiiragi, Yoshiki Matsuno, Takuya Ikura, Chang-geun Oh, Kazunori Fujimura, and Sin-ya Ayukawa.

# Two-channel Kondo Behaviors and Unconventional Phase transitions in Pr-based non-Kramers doublet systems

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In Pr-based non-Kramers systems with  $4f^2$  configurations, multipolar degrees of freedom in degenerated ground states play a key role in a variety of phase transitions and non-Fermi liquid (NFL) behaviors due to two-channel Kondo effect. It arises from interaction between the multipoles and two conduction electron bands. We report here possible multipole-driven phenomena of the non-Kramers doublets in a family of cubic Pr 1-2-20 systems and a hexagonal system PrRh<sub>3</sub>.

Pr 1-2-20 systems belong to the family of  $RT_2X_{20}$  (X = Al, Zn, and Cd) with the cubic CeCr<sub>2</sub>Al<sub>20</sub>type structure[1]. The ground states of  $Pr^{3+}$  under the crystalline electric field (CEF) are non-Kramers doublets having no magnetic dipole but active quadrupoles.[2]  $PrIr_2Zn_{20}$  exhibits an AFQ order at  $T_0$ = 0.11 K,[3] which is characterized by the quadrupoles aligned with the propagation vector of k =[1/2, 1/2, 1/2].[4] Furthermore, a superconducting transition occurs at  $T_c = 0.05$  K.[3] It was pointed out that the superconducting pair is mediated by the quadrupole fluctuations. For  $T > T_0$ , the magnetic specific heat  $C_{\rm m}$  follows  $-\ln T$  and the electrical resistivity  $\rho$  shows an upward curvature.[5] These peculiar temperature variations are reproduced by a two-channel Anderson lattice model, indicating formation of the quadrupole Kondo lattice in PrIr<sub>2</sub>Zn<sub>20</sub>.[6] If this is the case, a single-site quadrupole Kondo effect is expected to manifest when the  $Pr^{3+}$  ions are diluted with elements without 4f electrons.[7] Bearing this in mind, we have prepared single crystalline samples of the diluted Pr system Y(Pr)Ir<sub>2</sub>Zn<sub>20</sub>.[8] The measurements of specific heat and  $\rho$  showed that  $C_m/T$  follows  $-\ln T$  and  $\Delta \rho \propto \sqrt{T}$ , as predicted by the two-channel Kondo model.[7] Moreover, both data of  $C_{\rm m}$  and  $\Delta \rho$  per Pr ion are well scaled as a function of  $T/T_0$ , where  $T_0$  is a characteristic temperature of the two-channel Kondo effect. Therefore, we infer that the NFL behaviors result from the single-site quadrupole Kondo effect due to the hybridization of the  $4f^2$  states with two-channel conduction electrons.

On the other hand, a  $Pr^{3^+}$  ion in a crystallographic site with a hexagonal or trigonal point group has another type of non-Kramers doublets. The two-fold degenerated state has the magnetic dipole  $J_z$ and two components of the quadrupoles. The Pr atoms in the hexagonal PrRh<sub>3</sub> occupy two sites of the hexagonal 2*d* and trigonal 4*f* sites.[9] The magnetic susceptibility obeys the Curie-Weiss law with the paramagnetic Curie temperature  $\theta_p = +2.9$  K, which indicates ferro-type magnetic interaction between the Pr<sup>3+</sup> ions.[10] A cusp in C(T) appears at 3.0 K, whose peak broadens and shifts to higher temperatures as magnetic field is applied. The field dependence indicates a ferro-type magnetic order. The magnetic order. A broad tail of  $C_m$  was observed above  $T_C$ , which may result from short-range correlations and/or fluctuations of the active magnetic dipole and quadrupoles in the non-Kramers doublet. It is a striking contrast to an isostructural Kramers system NdRh<sub>3</sub> with 4*f*<sup>3</sup> configuration, where the magnetic dipoles of both the Nd sites simultaneously order at  $T_C = 5.0$  K.

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# Exploration for new layered superconductors with heavy elements

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Discovery of new superconductors with strong spin-orbit coupling has been desired. Our strategy for designing such new superconductors is to explore a layered structure which is composed of heavy elements. Since 2012, we have synthesized several BiCh<sub>2</sub>-based (Ch: chalcogen) layered compounds [1]. A typical BiCh<sub>2</sub>-based compound is composed of an alternate stacks of insulating (blocking) layers and electrically conducting BiCh<sub>2</sub> layers. Electron doping into the BiCh<sub>2</sub> layers make the system metallic, and superconductivity is observed in various electron-doped BiCh<sub>2</sub>-based compounds. However, optimization of local structure is also important to induce bulk superconductivity in this system [2]. In this presentation, we will show recent local structure analysis data of typical BiCh<sub>2</sub>-based system REO<sub>0.5</sub>F<sub>0.5</sub>BiS<sub>2</sub> [3]. In addition, the observation of unconventional isotope effect in a BiCh<sub>2</sub>-based superconductor will be discussed [4].

Based on the concept of the electron doping and the local structure optimization, we have designed new BiCh<sub>2</sub>-based superconductors and related compounds. We will introduce the crystal structure and physical properties of two BiCh<sub>2</sub>-based systems.

LaOBiSSe is a typical parent phase of BiCh<sub>2</sub>-based superconductors. A partial substitution of F for the O site induces electron carriers, and F-substituted  $LaO_{1-x}F_xBiSSe$  shows bulk superconductivity. We have investigated the effect of Ce substitution for the La site with the expectation of electron doping by the mixed-valence state of Ce.  $La_{1-x}Ce_xOBiSSe$  showed bulk superconductivity as the case of  $LaO_{1-x}F_xBiSSe$  [5]. Another doping method using the substitution of +4 ion for the La site will also be introduced in the presentation.

Electron carriers can be generated not only by the substitution of the blocking layer but also by insertion of rock-salt-type metal chalcogenide layers into a van-der-Waals gap of the BiCh<sub>2</sub>-based compounds. For example, PbS layer can be inserted into the van-der-Waals gap (interstitial position) of LaOBiS<sub>2</sub>. LaOBiPbS<sub>3</sub> is composed of La<sub>2</sub>O<sub>2</sub> blocking layers and Bi<sub>2</sub>Pb<sub>2</sub>S<sub>6</sub> conducting layers and shows a higher electron concentration than LaOBiS<sub>2</sub>. Using a similar strategy, AgBiS<sub>2</sub> layer can be inserted into the van-der-Waals gap of LaOBiS<sub>2</sub>, which results in the crystallization of La<sub>2</sub>O<sub>2</sub>Bi<sub>3</sub>AgS<sub>6</sub> [6]. La<sub>2</sub>O<sub>2</sub>Bi<sub>3</sub>AgS<sub>6</sub> shows a higher electron carrier density than LaOBiPbS<sub>3</sub> and exhibits metallic conductivity between 2 and 300 K. We discuss the possibility of superconductivity in those new layered chalcogenides.

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# Trial to Carrier-doping in Sr<sub>2</sub>IrO<sub>4</sub>/Ba<sub>2</sub>IrO<sub>4</sub>

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To realize the superconductivity in the Iridate system with the strong spin-orbit coupling will open the new world in the condensed matter physics. This is also one of the central issue in the "J-Physics". In particular,  $Sr_2IrO_4/Ba_2IrO_4$  has been predicted to be a high-temperature superconductor upon electron doping since it highly resembles the cuprate in crystal structure and magnetic coupling constant. Particularly, the remarkable resemblance between  $Sr_2IrO_4/Ba_2IrO_4$  and  $La_2CuO_4$  makes a good candidate to expect the unconventional HTSC in the  $Sr_2IrO_4/Ba_2IrO_4$ .

Indeed,

- 1) A low temperature STM study [1] on the K-doping (effectively election doping) in the clean surface of Sr<sub>2</sub>IrO<sub>4</sub> demonstrate the clear spin gap state.
- 2) Moreover, Y.K.Kim *et.al.* observed the low temperature nodal Fermi surface and high-temperature Fermi arcs[2]. These experimental results suggest the clear evidence of the d-wave pairing correlation. However, the direct evidence of the superconductivity such as zero resistivity (E=0) and Meisser effect (B=0) can not be observed in the bulk system. We have tried to observe the evidence of superconductivity in the carrier doped bulk material, however, not successful at present stage. Recently, we published our present experimental data in the following paper [3] [4].

In particular, Terashima *et.al.* demonstrated that the *d*-wave gapped state approaches the Fermi energy as the doped carrier increases, which show the striking similarity with those observed for underdoped cuprate, suggesting that superconductivity can be realized with increasing the carrier concentration.



Fig. shows the DC magnetic susceptibility of the  $Sr_{1.85}La_{0.15}$  IrO<sub>4-y</sub>F<sub>y</sub>, which shows the disappearance of the susceptibility on the F<sup>-</sup> doping.

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## Electronic structure of non-centrosymmetric system and Uranium compounds

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The electronic band structure and the Fermi surfaces have been calculated for cubic noncentrosymmetric compounds; PdBiSe, NiSbS, LaIrSi, Yb<sub>4</sub>Sb<sub>3</sub> and U<sub>3</sub>Ni<sub>3</sub>Sn<sub>4</sub> [1]. For some of them, the spin-structures are also investigated with R. Komiya. The electronic structure of other interesting systems, non-centrosymmetric CsW<sub>2</sub>O<sub>6</sub> and Mn<sub>3</sub>P, centrosymmetric LaNi<sub>2</sub>Cd<sub>20</sub>, Sn<sub>4</sub>P<sub>3</sub> are also investigated. Among them, the electronic structure UNi<sub>4</sub>B is reported here.

Recently, current-Induced magnetoelectric effect has been reported in a toroidal magnetic ordered state of UNi<sub>4</sub>B [2], where the uranium magnetic moment forms so called a ferro-toroidal ordering in a hexagonal lattice with  $T_N=20.4$ K [3]. However, now it is considered that UNi<sub>4</sub>B crystallizes in the orthorhombic structure with the symmetry # 63 (D<sub>2h</sub><sup>17</sup>, Cmcm) [4], rather than a hexagonal structure; #191 (D<sub>6h</sub><sup>1</sup> P6/mmm). Even in a hexagonal structure, uranium ions occupy two crystallographic sites (1a and 1b), so that two kinds of magnetic ions in different site could order with other transition temperatures. In the orthorhombic structure, uranium ions occupy 4 sites; two 4c and two 8c, so the situation is much complicated. B site is one in the hexagonal structure and four in the orthorhombic structure, so that a number of NQR signals is also a keto determine the structure.

The bandstructure calculations for UNi<sub>4</sub>B are investigated both with the hexagonal and the orthorhombic structure. The calculation for orthorhombic structure is very time consuming, so not yet completed. The density of states for the hexagonal structure is shown in Fig.1, in which it is found that the partial density of states for two uranium sites are very similar.



Fig. 1. The density of states for a hexagonal  $UNi_4B$ . The partial density of states for U(1a), U(1b), Ni(2d) and Ni(6i) are shown in magenta, blue, green and sky blue, respectively.

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## **Emergent electronic phenomena in hybrid f-/p-electron molecular materials**

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The occurrence of temperature- and pressure-driven valence transitions and giant negative thermal expansion effects in the mixed-valence rare-earth fullerene-based architectures with stoichiometry  $RE_{2.75}C_{60}$  [1] and the systematic tuning of the electronic response by both isovalent and aliovalent metal substitution are being systematically mapped out by a combination of structural (synchrotron X-ray diffraction) and spectroscopic (Raman spectroscopy, synchrotron X-ray absorption spectroscopy, resonant inelastic X-ray scattering) techniques at both ambient and elevated pressures (Fig. 1). Although these results are reminiscent of those of highly correlated Kondo insulators, the presence of the electronically active  $C_{60}$  sublattice, acting as an electron reservoir that accepts electrons from or donates electrons to the rare-earth 4f/5d bands under external stimuli, leads to a more complex electronic behavior.



Fig.1: (a) Pressure evolution of the unit cell volume and intensity ratio of  $\text{Sm}^{2+}$  and  $\text{Sm}^{3+}$  peaks in the XANES spectra of  $\text{Sm}_{2.75}\text{C}_{60}$ . (b) Unit cell volume *vs. x* in  $(\text{Sm}_{1-x}\text{Eu}_x)_{2.75}\text{C}_{60}$  and XANES spectra at the Sm  $L_3$ -edge of  $(\text{Sm}_{1-x}\text{Eu}_x)_{2.75}\text{C}_{60}$  and  $(\text{Sm}_{1-x}\text{Ca}_x)_{2.75}\text{C}_{60}$ . (c) Pressure evolution of the unit cell volume of  $(\text{Sm}_{2/3}\text{Eu}_{1/3})_{2.75}\text{C}_{60}$  and  $(\text{Sm}_{1/3}\text{Ca}_{2/3})_{2.75}\text{C}_{60}$ .

In addition, we identified an inhomogeneous orbitally selective (Jahn-Teller) anomalous metallic state with co-existing localized and itinerant electrons that is the parent of the highest- $T_c$  (38 K) and highest  $H_{c2}$  (>90 T, extremely strong binding of the Cooper pairs) unconventional fulleride superconductor [2]. This state appears to be harboring a hidden quadrupolar order [3], key to the understanding of the puzzling electronic properties of this state of matter.

Finally, polyaromatic hydrocarbons (PAHs) constitute a family of extended molecular carbon  $\pi$ -systems that can act as new electronic materials platforms beyond C<sub>60</sub>. Our systematic efforts unveiled that their intercalated compounds provide the first example of a 3D quantum spin-liquid state to 50 mK arising purely from  $\pi$ -electrons, while at the same time host orbitally entangled states, prerequisites of the emergence of quantum magnetism and exotic superconductivity [4].

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## Odd parity multipole ordering and off-diagonal response in 3d electron system

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A structure in which the local inversion symmetry is broken like zigzag chain and honeycomb structure with spin-orbit coupling are the key factor of realization of odd-parity magnetic multipole ordering. It is also expected that magnetoelectric effect [1,2], and optical magnetoelectric response like nonreciprocal directional dichroism. From this point of view, we conducted the following experiments to study the odd-parity magnetic multipole ordering and off-diagonal response in 3d electron system.

1) Nonreciprocal directional dichroism in chiral and polar magnets

The dynamical magnetoelectric effect appears as nonreciprocal dichroism of electromagnetic waves. We have investigate the magnetochiral dichroism of chiral antiferromagnet CsCuCl<sub>3</sub> and the directional dichroism in the magnetic resonance region of the polar magnetic material GaFeO<sub>3</sub>. Especially in CsCuCl<sub>3</sub>, magnetochiral dichroism in near infrared optical absorption is proportional to the magnetization, and the sign of magnetochiral dichroism depends on crystal chirality [3]. These responses are also useful as a detection of odd-parity magnetic multipole ordering.

2) Magnetoelectric effect and magnetoresistance in honeycomb antiferromagnets

Because the honeycomb structure breaks the local inversion symmetry, honeycomb antiferromagnets are candidate for the realization of odd-parity magnetic multipole ordering and magnetoelectric multiferroics. From this perspective, we investigated magnetoelectric effect in an antiferromagnetic insulator  $Co_4Ta_2O_9$  and magnetoresistance in an antiferromagnetic semiconductor  $CaMn_2Bi_2$ .

Co<sub>4</sub>Ta<sub>2</sub>O<sub>9</sub> belongs to the corundum-related structure with buckled honeycomb structure of magnetic Co<sup>2+</sup> ions [4]. This compound shows a large linear magnetoelectric effect in below  $T_N = 20$  K [5]. We reinvestigated magnetoelectric properties of Co<sub>4</sub>Ta<sub>2</sub>O<sub>9</sub> by using single crystal. Single crystal neutron diffraction measurement have revealed that the magnetic structure of Co<sup>2+</sup> spin moments are antiferromagnetically align in *ab* plane. In the magnetic ordered phase, both diagonal and off-diagonal terms of magnetoelectric effect are observed. Moreover, nonlinear magnetoelectric effect is observed, which is clearly different from similar compound Co<sub>4</sub>Nb<sub>2</sub>O<sub>9</sub> [6].

CaMn<sub>2</sub>Bi<sub>2</sub> has a trigonal CaAl<sub>2</sub>Si<sub>2</sub>-type structure [7]. This compound behaves semiconducting transport properties with small band gap.  $Mn^{2+}$  spin moments are antiferromagnetically ordered below  $T_N = 154$  K. We have investigated anisotropy of magnetization and magneto transport properties of CaMn<sub>2</sub>Bi<sub>2</sub> single crystal in pulsed high magnetic field. In the low temperature, this compound shows large magnetoresistance and the Hall plateau. The anomalous magneto-transport phenomena in this material may originates from Bi 6*p* band.

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## Search for New Ir- and Pt-Based Superconductors

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5d band metals of heavy elements such as Ir and Pt are promising candidate to show unconventional superconductivity caused by strong spin-orbit coupling of heavy 5d atoms, such as in Li<sub>3</sub>Pt<sub>3</sub>B [1] and SrPtAs [2]. The spin triplet pairing is reported to be dominant in the former, while the chiral *d*-wave or other unconventional pairings are theoretically predicted in the latter. Moreover, many Ir- and Pt-based superconductors have been discovered in recent years, indicating they are one of the hot spots for the search for novel superconductors. An example is ScIrP [3], which shows a superconducting transition at 3.4 K, accompanied by a moderately high upper critical field over 5 T at zero temperature.

We report our recent progress on the search for new Ir- and Pt-based superconductors. We prepared polycrystalline samples of platinum oxides, PtPt<sub>2</sub>O<sub>4</sub>, CaPt<sub>2</sub>O<sub>4</sub>, and NaPt<sub>3</sub>O<sub>4</sub>, and some ternary platinum pnictides. These three oxides show metallic behavior, which is quite rare for the platinum oxides, but the superconducting transition is not observed in electrical resistivity measurements down to 0.1 K. On the other hand, we find sharp drops of electrical resistivity to zero due to the superconducting transitions in the pnictide samples. We will present their superconducting and normal-state electronic properties.

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## **Anomalous Electronic Properties in Transition Metal Dichalcogenides**

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The layered MX<sub>2</sub> systems (M=transition metal elements, X=S, Se, and Te) show a variety of physical phenomena such as the charge density wave, superconductivity and so on [1]. Recent experimental studies on PdTe<sub>2</sub>, PtTe<sub>2</sub> and PtSe<sub>2</sub> with CdI<sub>2</sub>-type structure have indicated that these systems have new exotic electronic state, type II Dirac fermion state [2,3]. The results of angle resolved photoemission spectroscopy (ARPES) have revealed that the Dirac point exists at (0,0,k<sub>z</sub>) and the Dirac cone is strongly tilted along  $\Gamma$ -A direction (k<sub>z</sub>-direction) in these Pd and Pt dichalcogenides.

In the present work, we have performed the measurements of magnetoresistance and ARPES in NiTe<sub>2</sub> to clarify the electronic structure and confirm the existence of Dirac fermions in this system. NiTe<sub>2</sub> also has the CdI<sub>2</sub>-type structure and is a related system of the Pd and Pt dichalcogenides [1]. The single crystal of NiTe<sub>2</sub> was grown by Bridgeman technique. The magnetoresistance was measured using four-probe method in the magnetic field perpendicular to the Ni and Te layers. The ARPES experimental was carried out at BL-5U and 7U in UVSOR Facility, Institute for Molecular Science, Okazaki.

The band calculation in consideration of spin-orbit interaction has indicated that as well as Pd and Pt dichalcogenides, NiTe<sub>2</sub> is the type II Dirac fermion system with the Dirac point at  $(0,0,k_z)$  ( $k_z \sim 0.36c^*$ ). This compound shows the large linear magnetoresistance (~27% at 7T) at low temperatures, suggesting the existence of Dirac cone. The results of ARPES also suggested the existence of Dirac point at  $(0,0,k_z)$ . We measured the band dispersion along  $k_{//}$  ( $k_{//} \perp k_z$ ) at different  $k_z$ . At  $k_z=0$ , there are several hole- and electron-like bands around  $\Gamma$  and K points, respectively. With increasing  $k_z$ , the energy level of the hole-like band around  $k_{//}=0$  decreases, and the top of this band touches Fermi level at  $k_z \sim 0.37c^*$ . At this  $k_z$ , the band dispersion around  $k_{//}=0$  shows a linear  $k_{//}$ -dependence. This result has indicated that Dirac point exists very near Fermi level at  $(0,0,\sim0.37c^*)$  in NiTe<sub>2</sub>.

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## **Results of B01 group and search for superconductivity in f-electron systems**

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In B01 group, our aim is to study a variety of transport phenomena and novel quantum phases on the basis of the itinerant multipole. The 5f-electron systems are one of the main target to explore new materials and new phenomena, which are expected for unconventional superconductivity due to the itinerant multipole. Not only 5f-electron systems, but also new systems with d- and p-electrons are also our target. In particular, the strong spin-orbit coupling is a keyword in this system. First, We review our studies of B01 group in the past two years.

Next, we present our studies, focusing on unconventional/conventional superconductivity. In the ferromagnetic superconductor URhGe, we found that the field-reentrant superconductivity is quite sensitive to the uniaxial pressure along the b-axis in the orthorhombic structure. With increasing uniaxial pressure, the reentrant superconducting phase is lowered in field, and merges with the low-field superconducting phase. The spin reorientation field  $H_R$  decreases with the reentrant superconducting phase, indicating that the ferromagnetic fluctuations are tuned by the uniaxial pressure. The characteristic crystal structure with zigzag chains and the nearly hexagonal U atomic position play crucial roles for this field and uniaxial-pressure tuned ferromagnetic fluctuations.

We also present our recent studies on g-factor in URu<sub>2</sub>Si<sub>2</sub>, new superconductivity in f-electron systems, and Fermi surfaces in non-centrosymmetric heavy fermion U<sub>3</sub>Ni<sub>3</sub>Sn<sub>4</sub>.

## **Overview of Recent Studies in Our Group**

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In my presentation, I will overview three poster presentations submitted to this J-Physics workshop from our group.

The presentation given by M. Manago is about "NMR/NQR studies on ferromagnetic superconductor UCoGe under pressure". He has performed 59Co NMR and NQR measurements to reveal how the Ising-type ferromagnetic (FM) fluctuations are changed under pressure and are coupled to the superconductivity from the microscopic point of view. He found that the FM fluctuations at the SC transition temperature  $T_{SC}$  are strongly enhanced at 0.67GPa around the critical pressure  $P_c$  and starts to decrease at higher pressure 1.09 GPa. Since  $T_{SC}$  also shows a maximum around  $P_c$ , this result also supports the idea that the superconductivity is mediated by the FM fluctuations. The Knight shift was measured in the superconducting (SC) state at the pressure-induced paramagnetic (PM) side. He found that the Knight-shift decrease below  $T_{SC}$  is nearly unchanged in the field perpendicular to the *c* axis, and clarified that the superconductivity in the PM state is of spin triplet pairing.

The presentation by G. Nakamine is about "NMR study on the heavy-fermion (HF) superlattice (SL)". He has performed <sup>59</sup>Co-NMR measurements focused on the CeCoIn<sub>5</sub> block layers (BLs) in HF superconductor CeCoIn<sub>5</sub>/ normal-metal YbCoIn<sub>5</sub> SL and CeCoIn<sub>5</sub>/ HF antiferromagnet CeRhIn<sub>5</sub> SL to investigate how magnetic fluctuations in the several-layer-thick CeCoIn<sub>5</sub> BLs are changed by sandwich between YbCoIn<sub>5</sub> BLs or CeRhIn<sub>5</sub> BLs. He found that the low-temperature magnetic fluctuations of the CeCoIn<sub>5</sub> BL are weakened in a CeCoIn<sub>5</sub>/YbCoIn<sub>5</sub> SL by the Rashba spin-orbit effect but enhanced in a CeCoIn<sub>5</sub>/CeRhIn<sub>5</sub> SL by the magnetic proximity effect. In addition, he found that the magnetic properties are modified mainly at the interface region, suggesting that the interfacial interaction is the most important interaction in the SL compounds to tune the magnetic characteristics of the BLs

The presentation by S. Kitagawa is about "Possible Fulde-Ferrell Larkin-Ovchinnikov (FFLO) state realized in the S-type CeCu<sub>2</sub>Si<sub>2</sub>". He has performed <sup>63</sup>Cu – NMR / NQR measurements in order to investigate the magnetic and SC properties on a superconductivity dominant (S-type) single-crystal CeCu<sub>2</sub>Si<sub>2</sub>. He found the anomalous enhancement of  $1/T_1T$  just below  $H_{c2}(T)$  for magnetic field parallel or perpendicular to *c* axis. From the analogy of the results on  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub>, it is suggested that the FFLO phase might be formed in CeCu<sub>2</sub>Si<sub>2</sub>.

These studies have been performed in the collaboration with K. Deguchi, N. K. Sato, T. Yamamura, D. Aoki, M. Naritsuka, T. Ishii, T. Shibauchi, T. Terashima, Y. Kasahara, Y. Matsuda, H. S. Jeevan, C. Geibel, and F. Steglich.

# NMR study of URu<sub>2</sub>Si<sub>2</sub>

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In strongly correlated 5f-electrons systems, exotic electronic states appear due to the itinerant nature of 5f- electrons. Appearance of unconventional superconductivity under hidden ordered state in URu<sub>2</sub>Si<sub>2</sub> is typical example of it. In this study, the rotational symmetry of hidden ordered state and the superconducting gap symmetry are investigated by means of <sup>29</sup>Si NMR. As previously reported [1-3], the strength of 2-fold rotational breaking is weak and distributed in the basal plane of hidden ordered state. Consistently, the local 4-fold symmetry is supported via 4-fold Ruderman-Kittel interaction between the nearest neighbor Si sites determined by the NMR spin echo decay in the present study. The origin of muted 2-fold symmetry will be discussed. Concerning the superconducting gap symmetry, T-dependence of Knight shift along a-axis [4] and c-axis [5] indicates that the singlet paring (i.e. d-wave) is likely for the superconducting state of URu<sub>2</sub>Si<sub>2</sub>.

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# Non-Fermi liquid behavior in transport coefficients of U<sub>1-x</sub>Th<sub>x</sub>Be<sub>13</sub>

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Quantum criticality is a central topic of extensive current research interests in the strongly correlated electron systems because it is responsible for anomalous phenomena, such as non-Fermi liquid (NFL) behavior, observed in the vicinity of a quantum critical point (QCP). Moreover, it occasionally promotes the formation of unconventional superconductivity near a QCP.

UBe<sub>13</sub> undergoes superconducting (SC) transition at  $T_c \sim 0.8$  K from a NFL at zero field [1]. This feature is similar to that in other unconventional superconductors near a QCP in appearance but different in nature. For instance, UBe13 shows superconductivity before the formation of long-lived quasiparticles in the normal state [2,3]. This is in contrast to other unconventional superconductors, in which defined quasiparticles form the Cooper pairs even near a QCP while they are not so "well"defined as in an ordinary Fermi liquid. Moreover, distinct superconducting properties from those in other unconventional superconductors have been reported in UBe<sub>13</sub> [4,5]. In addition,  $U_{1-x}Th_xBe_{13}$ exhibits non-trivial enhancement of T<sub>c</sub> accompanied by an additional transition in the SC states for 0.02 < x < 0.04. Furthermore, it has been reported that the NFL behavior is even enhanced in the vicinity of x = 0.03. Namely, the SC transition temperature and the NFL behavior are most pronounced around x = 0.03. Thus, one of the key elements for understanding the exotic superconductivity in  $U_{1-x}Th_xBe_{13}$  is the clarification of the nature of the NFL behavior. While several theoretical studies on this issue have been done [6-8], the origin of the NFL behavior in  $U_{1-x}Th_xBe_{13}$ still remains unclear because of the lack of experimental results with high quality crystals. In our talk, we will report the recent progress of our transport measurements on high quality  $U_{1-x}Th_xBe_{13}$  and discuss the nature of the NFL behavior.

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## Characteristic Heavy Electron State in Sm*Tr*<sub>2</sub>Al<sub>20</sub> (*Tr*: transition metal)

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Heavy fermion (HF) compounds with cage structure have attracted much attention due to a variety of interesting phenomena, believed to be caused by enhanced *c-f* hybridization and small crystalline electric field splitting. Cage compounds with the general chemical formula  $RTr_2X_{20}(R)$ : rare earth, *Tr*: transition metal, *X*: Al, Zn, and Cd) crystallize in a cubic structure with the space group  $Fd\bar{3}m$ . Remarkable behaviors found in this series include an HF state with extremely large Sommerfeld coefficient of  $\gamma = 8 \text{ J/(mol} \cdot \text{K}^2)$  in YbCo<sub>2</sub>Zn<sub>20</sub> and a nonmagnetic Kondo effect and quadrupole fluctuation mediated superconductivity in Pr*Tr*<sub>2</sub>Al<sub>20</sub> (*Tr*: Ti and V).

In the Sm*Tr*<sub>2</sub>Al<sub>20</sub> family(*Tr*: Ti, V, Cr, and Ta), unusually field-insensitive phase transition [ $T_x$ =6.5 K(Ti), 2.9 K(V), 1.8 K(Cr), and 2.0 K(Ta)] and HF state have been discovered. The CEF ground state of these Sm compounds is expected to be a  $\Gamma_8$  quartet of the J = 5/2 multiplet. Since the size of ordered moment is extremely suppressed compared with that expected for a  $\Gamma_8$  ground state, the multipolar degrees of freedom included in the  $\Gamma_8$  state are expected to play some role in this field-insensitive ordered state. The magnetic susceptibility of the compound shows weak temperature dependences, which are evidently different from those for both free Sm<sup>2+</sup> and Sm<sup>3+</sup> ions. X-ray absorption spectroscopy (XAS) studies revealed that the Sm ions in the Sm $Tr_2Al_{20}$  family are in mixed valence states with an average Sm ion valence of 2.87 with no significant T dependence between 7 and 300 K[2,3]. These results suggest that valence fluctuation play an important role in the field-insensitive nature. We would like to discuss all these experimental results on Sm $Tr_2X_{20}$  and recent progress of theoretical works considering valence fluctuation [4].



Fig. (a) Magnetic susceptibility of  $\text{Sm}Tr_2X_{20}$ , (b) Sm valence determined by XAS vs Sommerfeld coefficient  $\gamma$  value on Sm-based compounds

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# Classification theory of odd-parity multipole order/superconductivity and electromagnetic responses

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Analysis of superconducting gap structure has been based on the group-theoretical classification by Sigrist and Ueda [1] for several decades. However, recent classification theory has revealed nontrivial gap structures beyond the Sigrist-Ueda theory. In this presentation, we show a comprehensive classification of " $j_z$ -dependent point nodes (gap opening)" [2]. Superconducting gap structure indeed depends on the angular momentum of Bloch states, in contrast to the prediction by the Sigrist-Ueda method. We suggest this unusual gap structure in heavy-fermion superconductors UPt<sub>3</sub>, UBe<sub>13</sub>, and PrOs<sub>4</sub>Sb<sub>12</sub>.

We also show a comprehensive group-theoretical classification of emergent multipole order in the condensed matter physics [3,4]. On the basis of the classification results in both real space and momentum space, we identify symmetry conditions and candidate materials for emergent electromagnetic responses. The responses include (1) piezoelectric effect, (2) magneto-piezoelectric effect, (3) Edelstein effect, (4) magneto-electric effect, (5) Antiferromagnetic spintronics, and (6) helical (FFLO) superconductivity.

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# Anisotropic quadrupole RKKY interactions under magnetic fields: ferroquadrupole order in PrTi<sub>2</sub>Al<sub>20</sub>

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Orbital orders in spin-orbital coupled systems have been attracted great attention due to their novel properties distinct from magnetic counter parts in recent years [1]. Pr-based 1-2-20 compounds are such systems with non-Kramers doublet ground state in the two f-electron crystalline electric field ground state. Among several compounds in the Pr-1-2-20 family,  $PrTi_2Al_{20}$  is believed to be a ferro-quadrupolar metal at low temperature [2]. A naïve expectation about the effects of magnetic fields on the ferro-quadrupole order based on a simple interacting quadrupole model [3,4] is that the  $3z^2-r^2$  type (so called O<sub>20</sub>) order smears out under magnetic fields parallel to [001] and it is thought to be trivial. However, experiments show a clear phase transition survives even under magnetic fields along [001] direction [5].

In order to explain the temperature-magnetic field phase diagram for  $PrTi_2Al_{20}$ , we point out in this talk that there exists field-induced interaction between quadrupole moments that can modify the order parameter configuration under magnetic fields. If this type of interaction is sufficiently large competing with the effective magnetic fields on the quadrupole moments, the first-order ferroquadrupole order remains even under finite magnetic fields. There, the quadrupole moments align along the direction *not* parallel to the effective magnetic field. Since such ordered-quadrupole moments are not induced by magnetic fields, this must be a phase transition. The analysis is based on the symmetry argument and we discuss the nature of possible phase transitions within the Landau theory. As a possible physical origin for this, the interaction mediated by conduction electrons is investigated on the basis of J = 3/2 electron system in a diamond structure [6].

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## Classification of multi-orbital superconductivity and its application

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Superconducting gap symmetry is one of the most important ingredients in unconventional superconductivity. In general, the gap symmetry is classified into irreducible representations of crystallographic point group, as summarized in a review paper by M. Sigrist and K. Ueda [1]. This has provided helpful information to analyse nodal structure in unconventional superconductors. However, since the discovery of iron-based superconductors, the importance of orbital degrees of freedom has been recognized. Indeed, orbital character of band electrons has a significant effect on the gap structure. A remarkable example is an exotic  $E_{2u}$  gap structure of UPt<sub>3</sub> obtained from the first-principles analysis [2]. It has shown in-plane twofold vertical line nodes on a small Fermi pocket mainly composed of the  $j_z = \pm 3/2$  orbitals. This gap structure seems to be an exception from a viewpoint of classification based on band electrons in the previous works, but the appearance is natural in the classification in total angular momentum j=5/2 space. Thus, it is important to consider the orbital character of band electrons in the classification.

First, we have studied superconducting gap structure in multi-orbital systems for three typical symmorphic group (O, D<sub>4</sub>, D<sub>6</sub>) from the group-theoretical classification [3]. We found that (i) a pairing state of  $\Gamma_9$  orbitals in D<sub>6</sub> possesses nontrivial momentum dependence. (ii) *s*-wave pairing states in degenerate orbitals cannot possess symmetry-protected but inevitable line nodes or gap minima. (iii) even the electron-phonon interaction can lead to non *s*-wave pairing states. As a prototypical example, we show that the *s*-wave gap structure in the BiS<sub>2</sub> layered superconductors can have twofold anisotropy on the Fermi surface [4], which may be consistent with the recent ARPES measurement [5].

Second, we have studied the gap structure in non-symmorphic magnetic group [6]. We have found that (i) in ferromagnetic superconductors like UCoGe, there appear inevitably horizontal line nodes on either a  $k_z=0$  plane or Brillouin zone (BZ) boundary perpendicular to  $k_z$  axis. (ii) in antiferromagnetic superconductors like UPd<sub>2</sub>Al<sub>3</sub>, an A<sub>g</sub>-type gap function possesses horizontal line nodes on the BZ boundary. In other words, the conventional fully-gapped *s*-wave superconductivity is forbidden, irrelevant to the pairing mechanism, as long as the Fermi surface crosses the BZ boundary.



Fig.1 (Left) Anisotropic pairing states driven by local pairing. (Right) Typical non-symmorphic crystal structures. Arrows on atoms represent the ordered magnetic moments.

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

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## Objective of the research project

In the present research project, we have revealed the three-dimensional (3D) electronic structure of uranium compounds by 3D ARPES which is a newly-developed experimental technique. In 3D ARPES experiment, all three parameters in ARPES experiments (two detecting angles of photoelectrons and incident photon energy) are entirely scanned, and complete volume data set which cover the entire Brillouin zone is obtained. The number of the ARPES spectra required to obtain 3D electronic structure is about  $5 \times 10^4$ , which is more than 15-fold of that of conventional ARPES measurements. We have established the data acquisition system which is capable of controlling the photoelectron analyzer, sample manipulator, and the beamline components of SPring-8 BL23SU.

#### Result

We have applied the 3D ARPES to the heavy Fermion superconductors URu<sub>2</sub>Si<sub>2</sub>. Figure 1 (a) shows the 3D Fermi surface map of URu<sub>2</sub>Si<sub>2</sub> in a paramagnetic phase. We found the intensity at the Fermi level is significantly enhanced at a small region in the Brillouin zone, and it is designated as 'hot spot'. The band structures parallel and perpendicular to the  $k_z$  direction in the vicinity of the hot spot are shown in Fig. 1(b). It was found that the hot spot is the contribution from narrow U 5*f* bands near the Fermi level, and the U 5*f* bands are strongly hybridized with Ru 4*d* bands in the vicinity of the hot spot. This suggests that U 5*f* states are strongly hybridized with Ru 4*d* states in URu<sub>2</sub>Si<sub>2</sub>. Furthermore, the overall 3D electronic structure was mostly explained by the band-structure calculation treating all U 5*f* electrons as being itinerant.

During the period of the research project, we have conducted the ARPES studies for  $UX_3$  (*X*=Al, Ga, and In) [1] and ThRu<sub>2</sub>Si<sub>2</sub> [2] in which elemental technologies of the 3D ARPES were utilized.

The work was conducted by the collaboration with Y. Takeda, T. Okane, Y. Saitoh, A. Fujimori, H. Yamagami, Y. Haga, E. Yamamoto, and Y. Ōnuki.



Fig. 1 The result of the 3D ARPES for  $URu_2Si_2$  in paramagnetic phase. (a) 3D Fermi surface maps and the location of 'hot spot'. (b) Band structures in the vicinity of the hot spot. Narrow U 5f bands are hybridized with dispersive Ru 4d states in the vicinity of the hot spot.

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## High Field Phases of URu<sub>2</sub>Si<sub>2</sub> and Related Compounds

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We have investigated the high field phases of URu<sub>2</sub>Si<sub>2</sub> and related compounds by means of neutron diffraction and X-ray circular magnetic dichroism. In URu<sub>2</sub>Si<sub>2</sub> and Rh dopes systems, a few distinct high field phases have been identified by magnetization measurements. In pure system, phases II-V appear in high magnetic fields. Three phases other than phase IV with saturated moment, magnetization shows non-saturated values close to 2/5, 1/2 and 3/5. A series of neutron diffraction experiments under pulsed magnetic fields shows that the magnetic phase is simplified by Rh doping and the phase II dominates. We have identified that the magnetic wave vector is commensurate [2/3,0, 0] for Rh 4 % sample[1]. More recently, the identical wave vector is observed for higher Rh doping[2], which is consistent with the trend of the phase diagram change by Rh doping. Moreover, the equivalence between the  $(1\ 0\ 0)$  and  $(0\ 1\ 0)$  was confirmed. When the Rh concentration is reduced, the commensurate [2/3, 0, 0] peak disappears at 2 % and in the pure system, incommensurate density wave like state is found[3]. These results indicate that the phase II in highly Rh doped system is different from the phase II of lightly doped system. It reminds us an open question that if there is a phase transition between these two phases along with Rh doping. We have investigated the details of wave vector distributions on pure compounds at around 2 K, to examine the difference between the phase II and phase V by using a white beam Laue method with a spallation source. The experimental results show that the wave vector is possibly not simple single value at low-temperature and that the distribution is quite sensitive to the temperature. A part of the complication arises from the magnetocaloric effect and the sensitiveness on the magnetic field sweep rate. It is confirmed by the direct sample temperature monitoring with a thermometer attached on the sample. We also note that no orthorhombic distortion is found within the resolution for the low field phase.

We have also examined the magnetic field induced meta-magnetic transition(crossover) in  $CeRu_2Si_2$  and  $CeRh_2Si_2$  by means of X-ray circular magnetic dichroism(XMCD) to understand the difference between U and Ce systems. In XMCD of  $CeRh_2Si_2$  with antiferromagnetic order at 38 K, a clear increase of XMCD signal of  $4f_1$  state is found for high magnetic fields. We also found an additional  $4f_0$  signal, which shows the admixture of itinerant state in ordered system of  $CeRh_2Si_2$ . As the second step, we examined  $CeRu_2Si_2$  with no magnetic order at zero field. By the success of recent technical development, we could measure XMCD at low temperature of 2 K, where the thermal effect on the meta-magnetic transition is much suppressed. We successfully found the induced enhancement of XMCD signal at the meta-magnetic transition. The XAS spectrum shows no obvious change of the valence state at the transition, which is consistent with the previous hard X-ray XAS measurement[4]. Another important finding is that the transition is quite gradual even at 2 K. It indicates that the transition is not a sharp  $1^{st}$  order transition, but a crossover like phenomena.

The present results shows the close relation between the wave vectors of high field phases and the nature of Fermi surface in URu<sub>2</sub>Si<sub>2</sub> system and the interplay between the itinerant and localized characters in related Ce compounds.

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# Direct Observation of Enhanced Paramagnetic-Limited Critical Magnetic Field in Electric-Field-Induced Superconductors

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Highly crystalline two-dimensional (2D) electron systems [1], which have been developed with the exfoliation and ionic gating techniques, provide us ideal platforms to explore the noncentrosymmetric superconductivity with strong antisymmetric spin-orbit coupling (ASOC). So far, we have demonstrated that the electric-field-induced superconductivity in SrTiO<sub>3</sub> [2] and MoS<sub>2</sub> [3] using electric double layer transistor (EDLT) configuration show the remarkably high in-plane critical magnetic fields  $H_{c2//}$  far beyond the usual Pauli paramagnetic limit of  $1.86T_c$ . The anomaly can be attributed to the spin-momentum locking or the spin-valley locking phenomena caused by the enhanced Rashba-type ASOC for SrTiO<sub>3</sub> EDLT [4] or the Zeeman-type ASOC with in-plane broken inversion symmetry for MoS<sub>2</sub> EDLT [3], which results in the enhancement of the paramagnetic limit. However, it has not been clearly confirmed whether the observed  $H_{c2//}$  is determined by the enhanced paramagnetic limit itself directly or by the orbital limit which can be measureable due to the enhancement of paramagnetic limit. In this presentation, we discuss our recent progress to answer this question through the detailed measurements of the critical field  $H_{c2}$  as a function of temperature *T* and angle  $\theta$  between the *c* axis and magnetic field direction.

The EDLT devices were fabricated at the atomically flat (001) surfaces of single crystals SrTiO<sub>3</sub> and MoS<sub>2</sub>, which were originally insulators before gating, by using an ionic liquid DEME-TSFI. In both cases, we found that  $H_{c2}(\theta)$  at low *T* does not obey the well-known 2D orbital limited behaviour (Tinkham's formula) accompanied with the cusp-like peak at  $\theta = 90^\circ$ , but shows rather the flattened peak, leading to the angular independent  $H_{c2}(\theta)$  in a narrow region around  $\theta = 90^\circ$ . In addition, we also observed that the *T* dependence of  $H_{c2//}$  deviates upward from 2D orbital limit formula,  $(1-T/T_c)^{1/2}$  law, at low *T*, which has been hardly reported in the orbital-limited superconductors. In SrTiO<sub>3</sub> EDLT, especially, we succeeded to obtain the further enhancement of  $H_{c2//}$  as compared with the previous study [2] by applying a higher gate voltage, resulting in  $H_{c2//}(T\rightarrow 0)$  exceeding  $6T_c$ . The above results can be the direct evidence for the observation of the Pauli paramagnetic limit enhanced by ASOC, leading to the unconventional superconducting properties, such as the helical and the mixed parity states.

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## **Characteristic Fermi Surface Properties in Cubic Compounds**

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We have studied Fermi surface properties of (1) AuAl<sub>2</sub>, AuGa<sub>2</sub>, and AuIn<sub>2</sub> with the CaF<sub>2</sub>-type structure, (2) CuS<sub>2</sub>, CuSe<sub>2</sub>, and (3) AuSb<sub>2</sub> with the FeS<sub>2</sub>-type structure, and (4) Pd<sub>3</sub>Bi<sub>2</sub>S<sub>2</sub> and (5) EuPtSi with the cubic chiral structure, via the dHvA experiments and the energy band calculations, as follows:.

1) We grew high-quality single crystals of AuAl<sub>2</sub>, AuGa<sub>2</sub>, and AuIn<sub>2</sub> by the Bridgman method. The Fermi surface and optical properties for three compounds were once studied from an interest of colors. The detected dHvA frequencies in the present study are found to be in a wide range of  $(0.1-13) \times 10^7$ Oe. The main dHvA branches for three compounds are in excellent agreement with the theoretical ones, but some dHvA branches with small dHvA frequencies are slightly deviated from the theoretical ones, especially in AuGa<sub>2</sub> and AuIn<sub>2</sub>[1].

2) CuS<sub>2</sub> and CuSe<sub>2</sub> were synthesized under high pressure of 4GPa and high temperature of 800 °C, revealing high-quality single crystal samples of RRR  $\simeq 100$ . Cyclotron effective masses are found to be in the range from 0.9 to 2.4  $m_0$ , which are mainly due to Cu-3*d* electrons (3/4-occupied  $e_g$  band). The superconducting upper critical fields  $H_{c2}(0)$  is small:  $H_{c2}(0) = 0.16$  kOe in CuS<sub>2</sub>( $T_{sc} = 1.5$  K) and  $H_{c2}(0) = 0.60$  kOe in CuSe<sub>2</sub>( $T_{sc} = 2.4$  K)[2].

3) We grew high-quality single crystals of  $AuSb_2$  by the Bridgman method. The Fermi surfaces of  $AuSb_2$  are found to be similar to those of NiSbS and PdBiSe with the ullmannite (NiSbS)-type cubic chiral structure because the crystal structures are similar each other and the number of valence electrons is the same between two different compounds. Note that each Fermi surface splits into two Fermi surfaces in NiSbS and PdBiSe, reflecting the non- centrosymmetric crystal structure[3].

4) In  $Pd_3Bi_2S_2$  with the cubic chiral structure, we detected four kinds of nearly spherical Fermi surfaces, where each Fermi surface splits into two Fermi surfaces. The splitting energy of main two Fermi surfaces is determined as 970 K[4].

5) EuPtSi crystallizes in the cubic chiral structure ( $P2_13$ , No. 198), which is the same as the one of MnSi with the skyrmion structure, and orders antiferromagnetically below a Néel temperature  $T_N = 4.05$  K. The magnetization at 2 K for the [111] direction indicates two metamagnetic transitions at the magnetic fields  $H_{A1} = 9.2$  kOe and  $H_{A2} = 13.8$  kOe and saturates above  $H_c = 26.6$  kOe. The present magnetic phase between  $H_{A1}$  and  $H_{A2}$  is most likely closed in the (H, T) phase diagram and is observed in a wide temperature range from 3.6 to 0.5 K. In this magnetic phase, we found giant additional Hall resistivity  $\Delta\rho(H)$  and magnetoresistance  $\Delta\rho(H)$ , reaching  $\Delta\rho(H) = 0.12 \ \mu\Omega \cdot cm$  and  $\Delta\rho(H) = 1.4 \ \mu\Omega \cdot cm$ , respectively[4,5].

We have collaborated with M. Kakihana, K. Nishimura, F. Suzuki, T. Yara, and Y. Ashitomi (Ryukyus), D. Aoki, A. Nakamura, and F. Honda (Tohoku), M. Nakashima and Y. Amako (Shinsyu), T. Sakakibara, S. Nakamura, and Y. Uwatoko (Tokyo), Y. Haga (JAEA), and T. D. Matsuda (Tokyo Metropolitan), together with H. Harima (Kobe).

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# Spin-Orbit-Coupled Ferroelectric Superconductivity

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In principle, metals cannot exhibit ferroelectricity because dielectric polarization is screened by conduction electrons. However, in 1965, Anderson and Blount predicted the existence of ferroelectric (FE) metals in which a FE-like structural transition occurs in the metallic state [1]. Since then, for half a century, a lot of experiments have been done to find the FE metal, and recently, a FE-like structural transition was discovered in metallic LiOsO<sub>3</sub> in 2013 [2]. More recently, following the discovery of FE metal, coexistence of superconductivity and ferroelectriclike order has also received a lot of attention. A candidate material of a ferroelectric superconductor, in which the FE-like order coexists with superconductivity, is SrTiO<sub>3</sub>. SrTiO<sub>3</sub> is a quantum paraelectric [3] whose dielectric constant is about 20,000 at low temperatures, but a pure compound does not exhibit ferroelectricity because the long-range FE order is suppressed by quantum fluctuation. On the other hand, SrTiO<sub>3</sub> exhibits ferroelectricity by substituting Sr with Ca or isotopic substitution of <sup>16</sup>O with <sup>18</sup>O. Furthermore, it becomes a metal by substituting Sr with La, Ti with Nb, or by removing oxygen. The carrier-doped SrTiO<sub>3</sub> becomes superconducting at a density as low as  $10^{17}$  cm<sup>-3</sup>. Based on these observations, Rischau *et al.* [4] performed various measurements for carrier-doped Ca-substituted SrTiO<sub>3</sub>. They mapped out a temperature-versuscarrier density phase diagram, which shows a coexistent phase of the superconductivity and the FE like order in the low carrier density regime. The above observations have generated a flurry of interest in the relationship between metallicity, ferroelectricity and superconductivity.

In the presentation, we will show results of our theoretical study about coexistence of the superconductivity and the FE-like order. We investigated the possibility of a FE superconductor based on the analysis of a model of two-dimensional Rashba superconductor. In our model, the effect of polar lattice displacement induced by a FE-like structural transition is included as a Rashba-type antisymmetric spin-orbit coupling. By calculating the free energy, in which the energy

loss of polar lattice displacement is included, we mapped out superconducting phase diagrams. It is shown that the FE superconducting state is stabilized under the applied magnetic field [Fig. 1], because of the suppression of the Pauli depairing effect in Rashba superconductors. Furthermore, it is revealed that the stability of the FE superconducting state is increased in the low carrier density regime, because of the Lifshitz transition at the crossing point of the Rashba spin-split bands. Therefore, the FE superconducting state can be stabilized without the applied magnetic field in the low carrier density regime.

#### $n = 0.3, y = 0.24, T_{c} = 0.043$ 1 0.8 SC Normal 7/T 0.6 0.4 SC + FE 0.2 0 0 0.5 2 2.5 1 1.5 $\mu_{\rm B}H_{\rm z}/T_{\rm c}$

Fig. 1. Superconducting phase diagram

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# Odd-parity multipole fluctuation and superconductivity in locally noncentrosymmetric system

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We investigate a two-sublattice Hubbard model with the sublattice-dependent antisymmetric spin-orbit coupling and Coulomb interaction on the basis of the random phase approximation. We approach to a nonsymmorphic and locally noncentrosymmetric structure; the crystallographic point group is  $D_{4h}$  and the local point group symmetry at specific ions is  $D_{2d}$ , lacking the  $\sigma_{v/d}$  mirror reflection symmetry, such as BaMn<sub>2</sub>As<sub>2</sub> and Sr<sub>2</sub>IrO<sub>4</sub>.

We find that a magnetic quadrupole, hexadecapole, and toroidal fluctuations appear by the antisymmetric spin-orbit coupling. The  $d_{xy}$ +p-wave state induced by these fluctuations is more suppressed by a modulate local parity violation than a globally inversion symmetry breaking. Our obtained gap function has a gapped/node structure protected by the nonsymmorphic symmetry in an inter-sublattice pairing. Furthermore, we obtain two triplet superconductivities, p+s-wave and p+d<sub>xy</sub>-wave states. The local parity violation prefers to these triplet states, in striking contrast to the globally inversion symmetry breaking.

## Magnetic properties of the new heavy fermion compounds Ce<sub>3</sub>TiBi<sub>5</sub>

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We have succeeded in growing the single crystals of hexagonal  $P6_3/mcm$  Ce<sub>3</sub>TiBi<sub>5</sub> by the Bi selfflux method [1]. Moreover the crystal structure of the newly discovered compound U<sub>3</sub>TiBi<sub>9</sub> was determined to be hexagonal  $P6_3/m$  [2]. Measurements of electrical resistivity, magnetic susceptibility, and specific heat have been made on the both compounds. The results indicate that both Ce<sub>3</sub>TiBi<sub>5</sub> and U<sub>3</sub>TiBi<sub>9</sub> are new compounds with an antiferromagnetic ordering temperature  $T_N$  of 5.0 and 31.5 K, respectively.

These crystal structures were only determined by the single crystal X-ray diffraction, because the both crystals are easily oxidizable in air. The crystal shape is very similar between the both crystals of Ce<sub>3</sub>TiBi<sub>5</sub> and U<sub>3</sub>TiBi<sub>9</sub>. The both crystal structures are hexagonal, although the space group is different. They have same Bi face-sharing octahedral chains centred of a Ti atom, however a Bi single chain or a Bi triangle prism lies between the three octahedral chains of Ce<sub>3</sub>TiBi<sub>5</sub> and U<sub>3</sub>TiBi<sub>9</sub>, respectively. Moreover the change from the single chain to the triangle prism additionally results in a slight octahedral chain's rotation about the c-axis [1, 2].

Figure 1 shows the temperature T dependences of the electrical resistivity  $\rho(T)$  and T derivative of the electrical resistivity  $\partial \rho / \partial T(T)$  for the (a) Ce<sub>3</sub>TiBi<sub>5</sub> and (b) U<sub>3</sub>TiBi<sub>9</sub>, respectively. The  $\rho(T)$ of the both compounds show the clear kink at  $T_N$  with decreasing T from room temperature. Each of these antiferromagnetic transitions was also observed on the T dependence of the magnetic susceptibility. And the both  $\partial \rho / \partial T(T)$  show a broad peak below the  $T_N$  in each.



Fig. 1. Temperature dependences of the electrical resistivity and temperature derivative of the electrical resistivity for the (a) Ce<sub>3</sub>TiBi<sub>5</sub> and (b) U<sub>3</sub>TiBi<sub>9</sub>, respectively.

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# Topological edge state of URu<sub>2</sub>Si<sub>2</sub> in superconducting state

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In URu<sub>2</sub>Si<sub>2</sub>, U atoms form body centred tetragonal lattice and show sublattice structure. Below approximately 0.75GPa and 17.5 K, the hidden ordered phase is realized. It has been shown that chiral d-wave superconductivity is realized in this phase. In k-space, the Fermi surface exists around the M point. Moreover, there are point nodes on north and south poles of the Fermi surface and horizontal line node on the surface  $k_z = 0$  [1,2]. Presence of point nodes is a characteristic feature of Weyl superconductivity. Weyl superconductor characterized by Weyl node, which are protected by a topological number [3,4].



Fig. 1. Relation between  $k_z$  and Chern number. There are some points where the Chern number changes. This result implies that the Weyl points exist there.

The Order parameter of URu<sub>2</sub>Si<sub>2</sub> in hidden ordered phase has not been uncovered. Hence, the hidden order parameter of URu<sub>2</sub>Si<sub>2</sub> has been experimentally investigated. Furthermore, the topological property of chiral d-wave superconducting state of URu<sub>2</sub>Si<sub>2</sub> is not clear. Therefore, we examined the topological property by calculating the Chern number and energy spectrum. First, we studied topological property by neglecting the influence of hidden order. For various magnitudes of the ratio of the inter-sublattice pairing and intra-sublattice pairing, we calculated Chern number and energy spectrum. As a result, we found that the Chern number changes at a certain momentum  $k_z$ . This changes of Chern number indicates that Weyl points exist at such  $k_z$ . Hence, we found such a new kind of Weyl point for a range of the ratio of pairing amplitudes. Moreover, we also investigated influences of hidden order.

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# Pressure-induced Phase Diagrams of the Chiral Magnet YbNi<sub>3</sub>Ga<sub>9</sub>: AC-Calorimetric Measurements up to 12 GPa

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The Yb-based compounds YbNi<sub>3</sub>X<sub>9</sub> (X = Al, Ga) crystallize in the trigonal ErNi<sub>3</sub>Al<sub>9</sub>-type structure with a chiral space group R32, making them unique candidates for systems exhibiting chiral magnetic structures as well as quantum critical phenomena [1, 2]. YbNi<sub>3</sub>Al<sub>9</sub> undergoes an antiferromagnetic (AFM) transition at  $T_M$  = 3.4 K [3]. By substituting 6% of Cu for Ni, a chiral soliton lattice (CSL) is realized under magnetic fields *B* applied parallel to the *a*-axis [1, 4, 5].

YbNi<sub>3</sub>Ga<sub>9</sub>, on the other hand, is an intermediate-valence system under ambient pressure [3]. We expect a pressure-induced magnetic order to occur in this compound. Indeed, an AFM order above  $P_c = 9$  GPa was inferred from the electrical resistivity  $\rho(T)$  and AC magnetic susceptibility measurements [2]. At  $P_c$ ,  $\rho(T)$  exhibits a linear-T dependence, i.e. non-Fermi liquid behavior.

In this work, we have investigated the quantum critical behavior and antiferromagnetic order in the chiral magnet YbNi<sub>3</sub>Ga<sub>9</sub> using AC-calorimetric measurements under pressures *P* up to 12 GPa. Thereby, magnetic fields *B* were applied parallel and perpendicular to the *c* axis. As shown in Fig. 1, the specific heat *C* divided by temperature *T*, *C*/*T*, dramatically increases with applying pressure. At 8.6 GPa  $\cong$  *P*<sub>c</sub>, *C*/*T* exhibits  $-\ln T$  dependence for 2 < T < 9 K. At T < 1.5K, *C*/*T* is saturated to a constant value 1 J/K<sup>2</sup>mol, i.e., Fermi-liquid behavior. At *P* = 9.3 GPa > *P*<sub>c</sub>, a broad maximum in *C*/*T* appears at 1.6 K, reflecting the AFM order. With increasing *P* further, the maximum shifts to higher temperatures and becomes a sharp  $\lambda$ -type peak at 5 K for *P* ≥ 11 GPa. By using the data of *C*(*T*, *P*) obtained under *B*⊥*c* and *B*//*c*, we have constructed *B*-*T* phase diagrams at each *P* value as shown in Fig. 2. The results at *P* ≥ 11 GPa have another field-induced ordered (FIOP) phase denoted by phase II appearing only for *B*⊥*c*. We discuss the origin of the FIOP in relation to the CSL for Yb(Ni<sub>1-x</sub>Cu<sub>x</sub>)<sub>3</sub>Al<sub>9</sub> and A-phase for MnSi [6] and EuPtSi [7].



**Fig. 1.** Specific heat divided by temperature C/T vs  $T^2$  for YbNi<sub>3</sub>Ga<sub>9</sub> under various pressures up to 12 GPa.



**Fig. 2.** *B*-*T* phase diagrams for  $B \perp c$  at 11.4 GPa and B//c at 11.5 GPa determined by specific-heat measurements.

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## Current-induced magnetization on CeRu<sub>2</sub>Al<sub>10</sub>

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Toroidal moment t is one of the parameters that describe a strength of the magnetoelectric coupling. Recently, S. Hayami *et al.* theoretically predicted that toroidal moment can be active also in metallic systems, and exotic phenomena such as magnetization induced by electric current can occur in the toroidal ordered metal [1].

In order to test this theory, we have performed magnetization measurements under electric current on an antiferromagnetic metal, UNi<sub>4</sub>B. We found that magnetization is induced by constant electric current in the ordered state of UNi<sub>4</sub>B [2]. Therefore, the validity of the theory is confirmed in part by the experiments. However, the fact that the magnetic structure of UNi<sub>4</sub>B is not fully confirmed makes it difficult to compare the experimental results with the theory, hindering further understanding of the observed phenomenon.

We have recently started searching for another candidate metal for the theory. In this presentation, we report another system which shows a current-induced magnetization, CeRu<sub>2</sub>Al<sub>10</sub>. It crystallizes into YbFe<sub>2</sub>Al<sub>10</sub>-type orthorhombic structure (symmetry: Cmcm,  $D_{2h}^{17}$ , No.63). Ce ions align along *c* axis forming zigzag structure. Ce ions participate an antiferromagnetic order at  $T_N = 27$  K. This order is characterized by a propagation vector q = (0, 1, 0) where magnetic moment align along *c* axis [3].

Single crystalline sample is grown by H. Tanida (Toyama Pref. Univ.). Large pieces of single crystalline are cut and polished into rectangular shape. Typical size is  $0.9 \times 1.9 \times 4.9 \text{ mm}^3$  Magnetization measurement under electric current for each setting of  $I \parallel a$  and c, and  $B \parallel a$ , b, and c have performed. We found magnetization is induced by electric current below  $T_N$  for the settings of  $(I \parallel a, B \parallel c)$  and  $(I \parallel c, B \parallel a \text{ and } b)$ . In the presentation, we are going to discuss about the cause of this phenomena based on the concept of odd parity multipoles.

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## Novel dual character of 4f electrons observed in Sm compounds

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We report a systematic study of Sm valence in the prototypical intermediate valence compound SmB<sub>6</sub>. Sm mean valence,  $v_{Sm}$ , was measured by the X-ray absorption spectroscopy as functions of pressure (1 < *P* < 13 GPa) and temperature (3 < *T* < 300 K). Pressure induced magnetic order (MO) was detected above  $P_c = 10$  GPa by resistivity measurements.

The estimated  $v_{\rm Sm} - T$  curves are shown in Fig. 1. The increase in  $v_{\rm Sm}$  with pressure and/or temperature indicates the localization of 4f electrons, analogous with the trend of Yb heavy fermion compounds which show a pressure induced nonmagnetic-magnetic transition. However  $v_{\rm Sm}$  at  $P_c$  in SmB<sub>6</sub> is anomalously far below a magnetic trivalency, which differs from the general case of nonmagnetic-magnetic transition in Yb and Ce compounds. According to the figure, the temperature dependence of  $v_{\rm Sm}$  is characterized by two features: (i) an almost temperature independent term seen in the higher temperature and higher pressure region, and (ii) a temperature dependent part in the rest region. For the latter, the  $v_{\rm Sm}$  -T curve depends on pressure as well. The deviation of  $v_{\rm Sm}$  from the trivalent state is therefore described as the sum of the temperature and pressure dependent term  $\delta v_{\rm Sm}(P,T)$  and the temperature independent term  $\Delta v_{\rm Sm}(P)$ :

 $v_{\rm Sm}(P,T) - 3 = \Delta v_{\rm Sm}(P) + \delta v_{\rm Sm}(P,T).$ 

Since  $\delta v_{\text{Sm}}(P,T)$  is easily suppressed by the thermal effects of a few hundred Kelvins, this term should be connected with the evolution of low-energy electronic correlations that can lead to the small modification of  $v_{\text{Sm}}$ . On the other hand,  $\Delta v_{\text{Sm}}(P)$  is hardly dependent on temperature up to at least 300 K, indicating that it is related to the valence fluctuations in higher-energy scheme through the large hybridization between f and conduction electrons. To understand this phenomenon, one may need to consider a peculiar mechanism such as proposed for U compounds, namely localized and delocalized f electrons coexist in multi f electrons system.



Fig. 1.  $v_{\rm Sm} - T$  curves at different pressures.

# Theory for Unconventional Non-Fermi Liquid Behaviors Observed in Ce<sub>x</sub>La<sub>1-x</sub>Cu<sub>5.62</sub>Au<sub>0.38</sub> (x=0.02-0.10)

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Quite recently, it was reported [1] that  $Ce_xLa_{1-x}Cu_{5.62}Au_{0.38}$  (x=0.02-0.10) exhibits a new type of quantum criticality in both magnetic and thermal properties, which is the same as those observed in a series of materials exhibiting quantum critical valence fluctuations (QCVF), such as  $\beta$ -YbAlB<sub>4</sub>, Yb<sub>15</sub>Al<sub>34</sub>Au<sub>51</sub>, and so on [2]. However, the temperature dependence in the resistivity  $\rho(T)$  at T < 0.5K is quite anomalous, i.e.,  $\rho(T) \propto (const. - T^n)$  with  $n \approx 0.75$  at x=0.05. This anomalous exponent is shown to be  $n=2(1-\zeta)$ , with  $\zeta$  being the weakly temperature dependent ( $0.5 < \zeta < 0.7$ ) critical exponent for the valence susceptibility due to the QCVF [3], provided that the impurities system of Ce ions exhibits the QCVF as in lattice systems as above. The observed critical exponent  $n \approx 0.75$  at x=0.05 is reproduced by choosing  $\zeta \approx 0.63$  which is consistent with the divergent behavior observed in the uniform magnetic susceptibility  $\chi \propto T^{-\zeta}$  [1,2].

This fact possibly sheds new light on the non-Fermi liquid properties observed in  $CeCu_{6-x}Au_x$  ( $x \sim 0.1$ ) at low temperature region which have been extensively discussed from the viewpoint of anomalous quantum magnetic transition [4]. This is because it has also been pointed out by the present authors [5,6] that the mother compound  $CeCu_6$  exhibits a rather sharp crossover in the valence of Ce ion under pressure [7] though it is a bit milder than the case of  $CeCu_2(Si,Ge)_2$  [8,9]. Indeed, a symptom of magnetic field and pressure induced quantum critical valence transition (QCVT) was reported [10], which had been predicted by the present authors [11,12].

The results described above is obtained by extending theoretical formalism of the QCVF to the impurities system, generalizing the theory [13] that explains an anomalous *T* dependence in the  $\rho(T) \propto (\text{const.} - T^{3/2})$  observed in a Kondo alloy La<sub>0.9</sub>Ce<sub>0.1</sub>Cu<sub>2.2</sub>Si<sub>2</sub> near the spin-density-wave quantum critical point [14,15].

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# Anisotropic ferro-quadupole interactions in quadrupole ordered system PrTi<sub>2</sub>Al<sub>20</sub>: Al-NMR and Magnetization studies

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The cubic compound  $\Pr Ti_2Al_{20}$  has the non-magnetic ground states of crystal field ( $\Gamma_3$ ). The  $\Gamma_3$  doublet have two pure electric quadrupoles of the  $\Gamma_3$ -type,  $O_{20} (3z^2 - r^2)$  and  $O_{22} (x^2 - y^2)$ , and magnetic octupole of  $\Gamma_2$ -type,  $T_{xyz} = (\sqrt{15} / 6) \overline{J_x J_y J_z}$ , where the bar represents the sum over possible permutation of the indices x, y, and z [1, 2]. The various measurements, such as specific heat, ultrasonic, neutron and NMR, reported that the ferro-quadrupole transition occurs near 2 K in this compound [3-6].

In this presentation, we will discuss the *H*-*T* phase diagrams from <sup>27</sup>Al-NMR and magnetization measurements in a single crystal of  $PrTi_2Al_{20}$ . When a magnetic field is applied along the <111> direction above 0.5 T, certain lines of the NMR spectrum split upon entering into the ordered phase, indicating breaking of the *C*<sub>3</sub> symmetry due to field induced magnetic dipole moment perpendicular to the field. This provides the first evidence for symmetry-breaking ferro-quadrupole order of *O*<sub>20</sub> type[6].

Recently, we observed the clear anomaly for the field-induced transition (the first transition) associated with the quadrupole moments near 2 T and 1.5 T for  $H \parallel <100>$  and <110> from NMR spectra and magnetization [7]. However, for  $H \parallel <111>$ , the absence of an anomaly occurs between 0.5 T and 11 T, suggesting the phase diagram has single ordered phase[6, 7]. The origin of anisotropic field induced transition is the competition between Zeeman interaction and ferro-quadrupole interaction coupled with the magnetic field. We will then present the detail of these experimental results and our calculation of Free energy for this system with mean field theory.

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# Multipole order of spin and orbitals in cobaltites

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Perovskite cobaltites are known to exhibit unusual magnetic and electronic phase changes due to the unique spin-state degree of freedom (total spin), which results from the competing Hund's coupling  $J_{\rm H}$  and crystal field splitting  $\Delta$ . Recently, it was found theoretically by considering correlation effects that exotic phases may emerge such as the Bose-Einstein condensation of excitons and spin-state crystalized phase due to spontaneous hybridization of the low-spin (LS) and high-spin (HS) or intermediate-spin (IS) states in cobaltites that are close to the spin-state crossover. The excitonic condensed state is nothing but a multipole order of spin and orbitals [1].

LaCoO<sub>3</sub> has LS (S = 0) insulator ground state. It transforms into magnetic insulator and then paramagnetic metal with increasing temperature, where the excited spin-states involved are controversial for half a century. On the other hand, in the recently found family of Pr-based cobaltites  $(Pr_{1-y}RE_y)_{1-x}Ca_xCoO_3$ , the ground state is non-magnetic insulator which becomes ferromagnetic metal at Tc with the first order phase transition with changes of many parameters of magnetic susceptibility, electronic conductivity, valence of Pr and Co ions and their spin-state. However, the order parameter driving the phase transition is still not known.

In the present study, high magnetic fields were used to finely tune the internal energies of excited spin-states in cobaltites and the spin-state change is followed using magnetization measurements up to 150 T with various temperatures. We have constructed *B*-*T* phase diagrams of LaCoO<sub>3</sub> and the Pr-based cobaltites. It was found that the ordered phases with low entropy emerge at high-magnetic fields in LaCoO<sub>3</sub> [2]. On the other hand, ferromagnetic metallic phase appears in Pr-based cobaltite when its insulator ground state is destroyed by magnetic fields [3].

The phase diagrams contrary to each other observed in LaCoO<sub>3</sub> and Pr-based cobaltites are understood in the following ways. In LaCoO<sub>3</sub>, magnetic field tunes the energy of excited HS or IS states so that it gets close to the LS state, where the spontaneous symmetry breaking takes place bringing about exotic phases such as the Bose-Einstein condensation of excitons and spin-state crystalized phase induced at megagauss fields [4]. On the other hand, in Pr-based cobaltite, high magnetic field breaks the Bose-Einstein condensation of excitons that already takes place at the ground state by separating the excited HS or IS states from the LS state [5].

To test above speculation for LaCoO<sub>3</sub>, we are working on measuring the spin-state evolution at ultrahigh magnetic fields beyond 200 T range to construct the full *B*-*T* phase diagram of LaCoO<sub>3</sub>, that is revealed only partially, yet. The magnetic field beyond 200 T is generated using electromagnetic flux compression technique in ISSP UTokyo. The spin-state evolution is monitored using specially developed high-speed 100 MHz magnetostriction measurement system based on fiber Bragg grating (FBG) technique [6]. Our measurement has been successful up to 250 T at 5 K, so far. We will present the up-to-date status of the measurement and discuss the physical significance of them.

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# Magnetic-Field-Induced Kondo Metal State by Closing the c-f Hybridization Gap in YbB<sub>12</sub>

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The specific heat and the magnetization of the Kondo insulator YbB<sub>12</sub> have been measured up to 60 T and 120 T, respectively. The specific heat experiment was conducted using a non-destructive conventional pulsed magnet. It is found that the Sommerfeld coefficient  $\gamma$  significantly increases at around 50 T where the insulator metal transition (MIT) occurs with a steep increase of the magnetization [1].  $\gamma$  reaches 67 mJ/(mol K<sup>2</sup>) at the high fields, which directly indicates that the quasiparticles possess heavy thermodynamic effective mass and the field-induced phase corresponds to a Kondo metal [2]. In Fig. 1, the obtained specific heat divided by temperature ( $C_p/T$ ) is shown as a two-dimensional (2D) image plot in *B-T* plane with a colour gradient scale. We also found that the field-induced Kondo metal has a rather high Kondo temperature around 200 K. This strong Kondo coupling proves that the energy gap collapse at the MIT does not correspond to the breakdown of the Kondo bound state. The steep increase of the magnetization at the transition manifests itself as a result of formation of the sharp density of states at the Fermi energy due to the Kondo resonance [3].

The magnetization process was measured up to 120 T using the single-turn coil technique [4]. A broad magnetization jump is observed at around 102 T in addition to the metamagnetism at the MIT around 50 T. Since the Kondo bound state must be broken when the magnetic field is strong enough, one of the possible origins for the magnetization jump is the transition from the Kondo metal to a normal metal. Such a transition is also expected in the theoretical calculation [2]. Similar successive two metamagnetic transitions are reported in anisotropic Kondo semimetal CeNiSn [5]; the effective mass of the itinerant quasiparticle is predicted to be enhanced between the two transition fields and reduced at higher magnetic fields. Although YbB<sub>12</sub> is regarded as isotropic Kondo insulator, similar two-gap structure is expected to be formed from the tight-binding theory [6]. Considering the anisotropic behaviour and possible mass reduction, the crystal field effect as well as the electronic correlation effect can be important to understand the high-field metallic phase of YbB<sub>12</sub>.



Fig. 1. 2D plot of the  $C_p/T$  in *B*-*T* plane with a colour gradient scale.

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# Superconducting point nodes depending on angular momentum *j*<sub>z</sub> ----Application to UPt<sub>3</sub>, UBe<sub>13</sub>, SrPtAs, etc.---

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Classification of a superconducting gap is one of the central subjects in the research field of unconventional superconductivity. Momentum dependence of the superconducting gap is closely related to symmetry of the superconductivity and the pairing mechanism. Since the superconducting gap structure can be identified by various experiments, combined studies of superconducting gap by theory and experiment may clarify the characteristics of superconductivity. Most of the theoretical studies have been based on the classification of order parameter by the crystal point group, which is summarized by Sigrist and Ueda (called Sigrist-Ueda method) [1]. However, their classification may not provide an exact classification of the superconducting gap.

For example, several studies have shown that the space group symmetry ensures the unconventional gap structures beyond the results of the Sigrist-Ueda method [2-5]. These theories classify not the order parameter, but the superconducting gap itself. Therefore, rigorous results are obtained for gap structures.

Especially regarding point nodes, many studies have discussed Weyl nodes in superconductors, namely point nodes protected by a nontrivial topological number [6-8]. However, there are only a few and less-known results about point nodes connected with crystal symmetry [2, 3].

In this study, we classify unconventional point nodes beyond the results of the Sigrist-Ueda method, by the group-theoretical analysis of the superconducting gap. We consider the gap structures on high-symmetry *n*-fold (n = 2, 3, 4, and 6) axes in the Brillouin zone, as examples of the symmetry-protected point nodes. Surprisingly, the analysis shows the existence of point nodes depending on the Bloch-state angular momentum  $j_z$  on a 3- or 6-fold axis. Furthermore, we suggest that such " $j_z$ -dependent point nodes" are realized in a heavy fermion superconductor UPt<sub>3</sub>. We also discuss superconducting gap structures in UBe<sub>13</sub>, SrPtAs, etc.

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### Magnetoresistance and Anomalous Hall effect in Weyl Magnet under rotated field

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As the first case in an antiferromagnet,  $Mn_3Sn$  is found to exhibit a large anomalous Hall effect, even at room temperature [1]. Usually, this anomalous Hall effect is known to be proportional to magnetization and thus has been observed only in ferromagnets. The spontaneous Hall resistivity in the antiferromagnet with vanishingly small magnetization indicates that the large fictitious field equivalent to a few hundred T must exist in the momentum space [1, 2]. Recently, we also reported a striking discovery of a large spontaneous Nernst effect of ~ 0.35  $\mu$ V/K in Mn<sub>3</sub>Sn at room temperature and ~ 0.6  $\mu$ V/K at low temperatures. The Nernst signals are found more than 100 times larger than what would be expected based on the magnetization [3]. Recent DFT calculation predicts that the large fictitious field may well come from a significantly enhanced Berry curvature associated with the formation of Weyl points nearby the Fermi energy  $E_F$  [4]. These larger anomalous Hall and Nernst effects observed in Mn<sub>3</sub>Sn are predicted to be derived from the large fictitious field enhanced by paired Weyl points in the band structure.

Here, we found strong experimental evidence of the Weyl fermions in Mn<sub>3</sub>Sn, namely, that the band structure is found consistent with DFT by ARPES and the chiral anomaly is clarified in the magnetotransport measurements. Generally, the chiral charge pumping between paired Weyl nodes can be derived in a magnetic field and an electric field applied parallel to each other and reduces the magnetoresistance with increasing the magnetic field. The negative magnetoresistance we observed is especially believed to be a signature of the chiral anomaly [5]. The recently identified candidates for topological semimetals, such as three dimensional topological Dirac semimetals Na<sub>3</sub>Bi [6] and Cd<sub>3</sub>As<sub>2</sub> [7] and inversion breaking Weyl semimetal TaAs [8], show peculiarly the negative magnetoresistance.

Thus, our experiments demonstrate that the large anomalous Hall and Nernst signals arise from the Berry curvature associated with the Weyl points near the Fermi energy [6]. In our talk, we also propose that the large anomalous Hall and Nernst effects are useful for memory and thermopile devices, respectively.

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# Inverse spin Hall effect in Mn-Sn amorphous alloy thin film

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Very recently, the non-collinear Kagome lattice antiferromagnet  $Mn_3X(X=Sn, Ge, Ga)$  alloys attract a lot of attention because of their large anomalous Hall and Nernst effect (AHE and ANE) while being an antiferromagnet with negligible magnetic moment [1-4]. The large AHE and ANE in  $Mn_3X$  is caused by the large fictitious field due to its Berry phase curvature in the momentum space. Since SHE/ISHE and AHE shares the same origin, the injection of pure spin current into these materials could potentially engender large ISHE or lead to exciting physics. Therefore, it is interesting to study the spin current injection into the Mn-X alloys.

In this work, we studied the inverse spin Hall effect in the  $\alpha$ -Mn and the Mn-Sn amorphous alloys. The spin current is generated in the ferromagnetic insulator YIG by the spin Seebeck effect, and then injects into the attached metal layer. Through a series of samples with different thicknesses, we obtained a sizable spin Hall angle ( $\theta_{SH}$ ) and long spin diffusion length ( $\lambda_{sd}$ ) for Mn, which are comparable with the previously reported values. Most importantly, we find that by doping Mn with 18% Sn, the  $\theta_{SH}$  of the amorphous non-magnetic Mn-Sn alloy is increased by as large as 20 times while the resistivity ( $\rho$ ) is reduced by about 10 times. Thus, the spin Hall conductivity ( $\sigma_{SH}$ ) of the Mn-Sn alloy is enhanced by 200 times comparing with pure Mn. The large enhancement is further corroborated by the spin pumping measurement. These results not only serve as essential references in studying the pure spin current phenomena in Mn-based alloys, but also offer a promising method in exploring future energy efficient spin Hall materials.

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# Study of the superconductivity of the ferromagnetic superconductor UCoGe under pressure

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Uranium-based ferromagnetic (FM) superconductors have been paid much attention because of these unusual feature that superconducting (SC) state arises inside the FM state [1]. UCoGe is a member of the FM superconductors, and it is pointed out that the Ising-type FM fluctuations are essential for the superconductivity [2]. This compound is a unique system in which the SC phase remains even if the FM state is suppressed by hydrostatic pressure.

We performed <sup>59</sup>Co NMR and nuclear quadrupole resonance (NQR) to reveal how the FM fluctuations are changed under pressure from the microscopic point of view. We found that the FM fluctuations at the SC transition temperature  $T_{SC}$  are strongly enhanced at 0.67 GPa around the critical pressure  $P_c$  and start to decrease at higher pressure 1.09 GPa. Since  $T_{SC}$  also gets maximum around  $P_c$ , this result also supports the idea the superconductivity is mediated by the FM fluctuations.

The measurements are also performed in the SC state at the paramagnetic (PM) side, and it was found that the Knight-shift decrease below  $T_{SC}$  is much smaller than the total spin part with the field perpendicular to the easy (*c*) axis, as shown in Fig. 1. This is a microscopic indication of the spin-triplet superconductivity in the PM side [3]. Nuclear spinlattice relaxation rate  $1/T_1$  exhibits node-like behavior with NQR without the magnetic field, and it seems that the gap structure is not so different from that in the FM side [4].

We will talk about the interpretation of the small change of the Knight shift in the SC state (Fig. 1) in the poster presentation. We will also discuss the order parameter in the PM SC state.



Fig. 1. <sup>59</sup>Co NMR Knight shift of UCoGe in the SC state with the field perpendicular to the *c* axis (33° away from the *b* axis to the *a* axis) under 0.67 GPa (PM state). The change of the Knight shift below  $T_{SC}$  is much smaller than the spin part  $K_{spin}$  in any measurement fields.

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# Study of Magnetic Structure in SmBe13 with Broken Local-Inversion Symmetry

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The intermetallic compounds MBe<sub>13</sub> (M = rare earths and actinides) crystallize in a NaZn<sub>13</sub>-type cubic structure with the space group  $Fm\bar{3}c$  (No. 226,  $O_h^6$ ), where *M* atoms are located at the 8*a* site without local-inversion symmetry (*O*). [1] Among them, UBe<sub>13</sub> is well known as a heavy-fermion superconductor, and a number of studies have been performed in order to unveil the origin of its unconventional superconductivity as well as non-Fermi-liquid behaviour. [2] On the other hand, it has been revealed from previous neutron scattering measurements that most heavy-rare-earth MBe<sub>13</sub> compounds undergo a helical-magnetic order with propagation vector of  $Q \sim <0, 0, 1/3>$  at low temperatures, where magnetic moments lie in the (001) plane. [3] Although light-rare-earth MBe<sub>13</sub> compounds are also expected to exhibit similar helical-magnetic order, there is no microscopic study on their magnetic structures thus far.

We have recently succeeded in growing single crystalline samples of SmBe<sub>13</sub> by the Al-flux method, and constructed a detailed magnetic phase diagram from several bulk measurements. [4] The magnetic phase diagram of this compound consists of three regions (named low-*B*, high-*B* and low-*T*, and high-*B* and high-*T* phases) below the ordering temperature  $T_M \sim 8.3$  K. In the present study, we performed <sup>9</sup>Be NMR measurements on single crystalline SmBe<sub>13</sub> at the magnetic field *B* of ~ 2 T and 5 T in order to investigate the magnetic structure of each phase microscopically. In the low-*B* phase ( $B \sim 2$  T), we found a clear difference in the obtained NMR spectra at 1.8 K between B // <001>and <110>, as shown in Fig. 1. These obtained spectra cannot be explained by the helical magnetic structure reported in the heavy-rare-earth MBe<sub>13</sub> systems. The most plausible helical structure for reproducing the obtained spectra is that the propagation vector is same as that observed in the heavyrare-earth MBe<sub>13</sub> systems, namely Q = <0, 0, 1/3>, but the magnetic moments lie in the (111) plane. In addition, NMR spectrum at T = 2 K and  $B \sim 5$  T suggests that uncompensated "up-up-down"-type antiferromagnetic order occurs in the high-*B* and low-*T* phase.

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Fig. 1. <sup>9</sup>Be NMR spectra of SmBe<sub>13</sub> at T = 1.8 K in the low-*B* phase ( $B \sim 2$  T), measured in external fields parallel to (a) <001> and (b) <110>.

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### Symmetry analysis of electrical switching of antiferromagnet

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Antiferromagnetic states are insensitive to electric or magnetic perturbations, since it has no net electric or magnetic polarizations. Several approaches have been proposed to operate the antiferromagnetic states [1, 2]. Especially, the switching scheme of current-induced antiferroic spinorbit torque recently attracted much attention in the field of antiferromagnetic spintronics [3]. This scheme has been proposed by the theoretical investigation and conducted by the celebrated experiment of Wadley *et. al* [4].

In the presentation, we conduct the symmetry analysis of current-induced AFM switching. The antiferroic (Neel) spin-orbit torque is derived from the local non-centrosymmetric property of crystal structure as the previous theoretical studies clarified [4, 5, 6]. With the use of representation theory and magnetic point group, we identify what "antiferromagnetic order" can be switched by applying electric current. Although previous studies have focused on the simple antiferromagnets [6], our symmetry analysis can be applied to the systems with complex magnetic configurations. Following the symmetry analysis, we propose the candidates for the switchable antiferromagnets and also discuss the relation with the ferro-toroidic order and the possibility to observe it.



Fig. 1. The crystal structure and the magnetic structure of CuMnAs [4]. The electric current along the x- or y-direction induce the corresponding antiferromagnetic order.

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# Theory of superconductivity in hole-doped monolayer MoS<sub>2</sub>

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The layered transition metal dichalcogenides (TMDs) have attracted much interest as a key semiconducting material, not only for electrical and optoelectronic devices but for spintronic devices. Moreover, *electron-doped* monolayer MoS<sub>2</sub> exhibits superconductivity, whose in-plane upper critical field  $H_{c2}$  is much larger than the Pauli limit [1]. The relatively high  $H_{c2}$  may be due to the broken spatial inversion symmetry that favors spin-valley locking [2].

From the electronic band structure point of view, superconductivity in *hole-doped* monolayer MoS<sub>2</sub> is expected to exhibit much variety of Cooper pairs since the valence bands are composed of 3 *d*-electron orbitals. Namely, the spin splitting in valance bands (VB) near the



Fig. 1. Orbital and spin components of the valence bands near Fermi level with the relevant Cooper pairs. The hexagon represents the Brillouin zone.

Fermi level is about 10 times larger than that of conduction bands (CB) [3] as shown in Fig. 1. Although a realization of superconducting state has not been reported in hole-doped monolayer  $MoS_2$  [4], we theoretically explore the Cooper pair symmetry to be realized in hole-doped monolayer  $MoS_2$ .

We investigate the 3-orbital tight-binding model, which is deduced from the first-principle band calculation [3]. We also introduce the atomic spin-orbit interaction (SOI), and assume the isotropic on-site attractions as summarized in Tables 1 and 2. We solve the linearized BCS gap equation, in which the complicated gap functions are decomposed into the spin-singlet  $D_0$  within  $|0\rangle$  orbital, spin-singlet and orbital-triplet **D**, and spin-triplet and orbital-singlet **d** in  $|\pm 2\rangle$  orbitals as follows:

$$\Delta_{nn'}^{\sigma\sigma'} = -\frac{1}{N} \sum_{k} \sum_{mm'} \langle nn' | \hat{g} | mm' \rangle \langle \hat{c}_{-km'\sigma'} \hat{c}_{km\sigma} \rangle, D_0 = \Delta_{00}^{\uparrow\downarrow}, D_z = \frac{1}{2} \left( \Delta_{+2-2}^{\uparrow\downarrow} + \Delta_{-2+2}^{\uparrow\downarrow} \right), d_z = \frac{1}{2} \left( \Delta_{+2-2}^{\uparrow\downarrow} + \Delta_{+2-2}^{\downarrow\uparrow} \right)$$

Figure 2 shows the doping (x=4-n, *n* being the electron density per unit cell) dependence of  $T_c$  and the ratio of the Cooper-pair components for U=0.5,  $J_0=J_2=-U/2$ , and  $\lambda =0.1$ [eV]. At low dopings,  $d_z$  dominant pairing is realized due to the hole pockets near the K and K' points ( $|\pm 2\rangle$  orbitals). For further doping above x=0.025, the hole pocket near the  $\Gamma$  point ( $|0\rangle$  orbital) appears, leading to the enhancement of  $T_c$  and the ratio of  $D_0$  and  $D_z$  as compared with  $d_z$ .

	xy	$x^2 - y^2$	$Z^2$		
xy	U	$U - 2J_2$	$U-2J_0$		
$x^2 - y^2$	$U - 2J_2$	U	$U - 2J_0$		
$Z^2$	$U - 2J_0$	$U-2J_0$	U		
Table 1. Direct attractions $\langle uv   \hat{a}   uv \rangle$					

	xy	$x^2 - y^2$	$Z^2$		
xy		$J_2$	$J_0$		
$x^2 - y^2$	$J_2$		$J_0$		
$z^2$	$J_0$	$J_0$			
Table 2. Exchange attractions $\langle \mu \nu   \hat{g}   \nu \mu \rangle$					



Fig. 2. Doping dependence of  $T_c$  and the ratio of the components of the Cooper pairs for U = 0.5,  $J_0 = J_2 = -U/2$ ,  $\lambda = 0.1$  [eV].

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# Microscopic Description of Electric and Magnetic Toroidal Multipoles in Hybrid Orbitals

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Mutual interplay between fundamental degrees of freedom of electrons in solids, i.e., charge, spin, and orbital, has attracted growing interest in various context. The concept of multipole has been developed to describe such complex electronic degrees of freedom, such as magnetic monopole and electric quadrupole, in a unified manner [1-3]. Under the space-time inversion group, there are four types of multipoles according to their presence/absence of spatial inversion and time-reversal symmetries: electric (E) multipole, magnetic (M) multipole, magnetic toroidal (MT) multipole, and electric toroidal (ET) multipole. Among the toroidal multipoles, the MT dipole has been extensively investigated due to its potential role for exotic phenomena, such as magneto-electric effect and nonreciprocal directional dichroism [4]. Such a MT dipole is often identified with a vortex-type magnetic orderings over several atomic sites as a classical object. However, MT multipole can arise even at each atomic site as a quantum object. Thus, it is desirable to obtain quantum-mechanical operator expressions of toroidal multipoles, in contrast to previous discussions as classical electromagnetic quantities [5]. Once we obtain such expressions, we can clarify when it can be a primary order parameter characterizing thermodynamic phases in condensed matter.

In the present study, we discuss a general microscopic formalism to describe not only MT but also ET multipoles [6]. By summarizing the classical description in the expansion of electromagnetic potentials, and using the mutual correspondence among four fundamental multipoles, we derive the quantum-mechanical operator expressions of both ET and MT multipoles. We demonstrate that the atomic ET and MT multipoles can be activated in the Hilbert space spanned by orbitals with different azimuthal quantum number, e.g., *s-d*, *p-d*, and *d-f* hybrid orbitals, as shown in Fig. 1. We find that the atomic ET and MT multipoles in addition to ordinary E and M multipoles constitute a complete set to express an arbitrary degree of freedom in the hybrid orbitals. We also demonstrate emergent cross-correlated couplings, such as magneto-electric and magneto(electro)-elastic couplings, in the presence of an ET or MT multipole ordering.

basi	s	$\mathcal{P}$	l = 0	l = 1	l = 2	l = 3	l = 4	l = 5	l = 6
<b>s-s</b>	(1)	+	Е	-	-	4	_	-	-
p- $p$	(9)		Е	М	E	-	-	-	-
d- $d$	(25)		Е	Μ	E	Μ	Е	-	-
f- $f$	(49)		E	Μ	Е	М	Е	Μ	E
s- $d$	(10)	+	-	-	E/MT	-	-	-	-
p- $f$	(42)		-	-	E/MT	M/ET	E/MT	- 4	-
s-p	(6)	-	-	E/MT	-	-	-	-	-
s- $f$	(14)		-	-	-	E/MT	-	-	-
p- $d$	(30)		-	E/MT	M/ET	E/MT	-	-	-
d- $f$	(70)		-	E/MT	M/ET	E/MT	M/ET	E/MT	-

Figure 1. Multipoles activated in non-hybrid (intra) and hybrid (inter) orbitals. The number of independent multipoles to span the relevant Hilbert space is indicated in the parenthesis.  $\mathcal{P}$  represents the parity of the active multipoles with the rank *l*.

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Thermoelectric properties of one dimensional telluride  $M_4$ SiTe<sub>4</sub> (M = Ta, Nb)

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Thermoelectric cooling is a promising candidate for the next-generation refrigeration technologies in replacing vapor compression cooling using gaseous refrigerants. At present, Peltier devices using Bi<sub>2</sub>Te<sub>3</sub>-based materials have been put to practical use at around room temperature. Practical cooling at much lower temperatures enables us to refrigerate various cryogenic electronics devices, resulting in much wider applications of thermoelectric cooling.

Here, we report one-dimensional telluride  $M_4$ SiTe<sub>4</sub> (M = Ta, Nb) and its substituted compounds show high thermoelectric performance at low temperature[1][2]. Ta<sub>4</sub>SiTe<sub>4</sub> and Nb<sub>4</sub>SiTe<sub>4</sub> were first synthesized by Badding *et al.*, in a needle like crystalline form[3][4]. We prepared whisker crystals with typically several mm long and several µm in a diameter and measured their thermoelectric power and electrical resistivity. Thermoelectric power of Ta<sub>4</sub>SiTe<sub>4</sub> and Nb<sub>4</sub>SiTe<sub>4</sub> whisker crystals reaches  $S = -400 \ \mu V \ K^{-1}$  and  $S = -200 \ \mu V \ K^{-1}$  at 150-200 K, respectively, while maintaining low resistivity of  $\rho \sim 2 \ m\Omega$  cm as shown in the figure. These yield power factor  $P = S^2/\rho \sim 80 \ \mu W$ 

cm<sup>-1</sup> K<sup>-2</sup> of Ta<sub>4</sub>SiTe<sub>4</sub> and  $P \sim 25\mu W \text{ cm}^{-1} \text{ K}^{-2}$  of Nb<sub>4</sub>SiTe<sub>4</sub> at optimum temperature of 130 K, which are larger than and comparable to ~ 35  $\mu$ W cm<sup>-1</sup> K<sup>-2</sup> of Bi<sub>2</sub>Te<sub>3</sub>-based materials at room temperature, respectively. The first principles calculation results suggest that the high thermoelectric performances in  $M_4$ SiTe<sub>4</sub> caused by the coexistence of а one-dimensional electric structure and a very small spin-orbit gap at the Fermi energy and the larger gap in Ta<sub>4</sub>SiTe<sub>4</sub> than that in Nb<sub>4</sub>SiTe<sub>4</sub> due to the stronger spin-orbit couping of Ta than that of Nb gives to rise to the difference of the thermoelectric performance between them.



Fig. Electrical resistivity (left axis) and thermoelectric power (right axis) of Ta<sub>4</sub>SiTe<sub>4</sub> and Nb<sub>4</sub>SiTe<sub>4</sub> whisker crystals.

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# FFLO state in CeCu<sub>2</sub>Si<sub>2</sub> revealed by Cu-NMR

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The first heavy-fermion superconductor  $CeCu_2Si_2$  discovered in 1979[1] was considered to be a nodal unconventional superconductor from the experiments on polycrystalline samples[2] However, recent specific heat[3] and thermal conductivity measurements[4] on a superconductivity dominant (S-type) single crystal of  $CeCu_2Si_2$  down to 40 mK strongly suggested that  $CeCu_2Si_2$  possesses more than two full gaps, the size of which are quite different with each other. In addition, it was shown that the superconductivity of  $CeCu_2Si_2$  is robust against the impurities introduced with an electron irradiation, which is contradict with the  $s_{+-}$ -wave model but consistent with the multi-band  $s_{++}$  wave model. At present, the identification of the SC-gap symmetry is an important issue in  $CeCu_2Si_2$ .

As for the parity of the superconductivity, the decrease of the Knight shift in the SC state[5] and the strong suppression of  $H_{c2}(T)$  in any magnetic field direction[6] indicate a spin-singlet pairing. The strong suppression of  $H_{c2}(T)$  indicates that the Pauli pair-breaking effect seems to be dominant. However, the clear first-order phase transition has not been observed, but it was suggested that the multiband character of superconductivity may suppress the first-order transition[6]. The conditions for the realization of the FFLO state seems to be satisfied in CeCu<sub>2</sub>Si<sub>2</sub>, but an experimental evidence of the FFLO state has not been reported so far. This is because experimental probes to detect the FFLO state is limited.

We have performed <sup>63</sup>Cu-nuclear magnetic resonance (NMR)/nuclear quadrupole resonance (NQR) measurements in order to investigate the magnetic and SC properties on a S-type single-crystal CeCu<sub>2</sub>Si<sub>2</sub>[7].  $1/T_1$  at zero field is almost identical to that in the previous polycrystalline samples down to 130 mK and slightly deviates downward from that below 120 mK.  $1/T_1$  in the SC state can be fit with the two-gap s+--wave rather than the two-gap s++-wave model down to 90 mK. Under magnetic fields, the spin susceptibility in both directions clearly decreases below  $T_c$ , indicative of the formation of the spin singlet pairing. The residual part of spin susceptibility is well understood by the field induced residual density of states evaluated from  $1/T_1T$ , which is ascribed to the effect of the vortex cores. In addition, we found the anomalous enhancement of  $1/T_1T$  just below  $H_{c2}(T)$  for magnetic field parallel or perpendicular to *c* axis. From the analogy of the results on  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub>[8], we suggests that the FFLO phase is formed in CeCu<sub>2</sub>Si<sub>2</sub>.

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### Non-Fermi Liquid Behaviors in Pr 1-2-20 Systems

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Non-Fermi liquid (NFL) behaviors in *f*-electron systems accompany quantum criticality where the quantum fluctuations are strongly enhanced due to the competition between the on-site magnetic Kondo effect and the inter-site Ruderman-Kittel-Kasuya-Yosida interaction [1]. On the other hand, another type of the NFL state due to quadrupole Kondo effect was proposed for some U-based systems where the local quadrupoles of  $5f^2$  electrons are over-screened by the two-channel conduction bands [2]. However, it is still controversial because crystalline electric field (CEF) levels and valences of the U ions are not well defined. Thereby, we have focused on cubic Pr 1-2-20 systems with  $4f^2$  configuration to reveal the characteristic NFL behaviors due to the quadrupole Kondo effect [3].

Pr 1-2-20 systems belong to the family of  $RT_2X_{20}$  (X = Al, Zn, and Cd) with the cubic CeCr<sub>2</sub>Al<sub>20</sub>type structure [4]. The ground states of Pr<sup>3+</sup> under the CEF are non-Kramers doublets having no magnetic dipole but active quadrupoles [3]. PrIr<sub>2</sub>Zn<sub>20</sub> shows the antiferroquadrupole (AFQ) order at  $T_Q = 0.11$  K [5]. Furthermore, a superconducting transition occurs at  $T_c = 0.05$  K. Thereby, it was pointed out that the superconducting pair is mediated by the quadrupole fluctuations. For  $T > T_Q$ , the magnetic specific heat  $C_m$  follows  $-\ln T$  and the electrical resistivity  $\rho$  shows an upward curvature [6]. These peculiar temperature variations are reproduced by a two-channel Anderson lattice model, indicating formation of the quadrupole Kondo lattice [7].

To reveal the interplay between the NFL behavior and the *c*-*f* hybridization in PrIr<sub>2</sub>Zn<sub>20</sub>, we have studied the effect of Ga and Cd substitutions for Zn on the NFL behavior by the measurements of the specific heat *C* and  $\rho$  of PrIr<sub>2</sub>Zn<sub>20-x</sub>Ga<sub>x</sub> ( $0 \le x \le 0.25$ )[8] and PrIr<sub>2</sub>Zn<sub>20-y</sub>Cd<sub>y</sub> ( $0 \le y \le 0.3$ ). With increasing *x* and *y*, a characteristic temperature *T*<sub>0</sub> of the two-channel Kondo effect defined as the temperature where the magnetic entropy *S*<sub>m</sub> reaches (3/4)*R*ln2 is increased by a factor of 3.5 and 1.7 for *x* = 0.25 and *y* = 0.3, respectively. The increments of *T*<sub>0</sub> are attributed to the possible split of the ground state doublet by lowering of symmetry at the Pr site. On the other hand, we define another characteristic temperature *T*<sub>R</sub> below which  $\rho(T)$  starts decreasing with the upward curvature. *T*<sub>R</sub> increases with *x* by a factor of 1.2 for *x* = 0.25, while *T*<sub>R</sub> decreases with *y* by a factor of 0.6 for *y* = 0.1. The opposite variations of *T*<sub>R</sub> for the Ga and Cd substitutions may result from the opposite change in the strength of the *c*-*f* hybridization. Thereby, in the Ga substituted system, the quadrupole Kondo lattice is stabilized by the enhanced *c*-*f* hybridization due to the increment of the 4*p* electronic density.

On the other hand, a single-site quadrupole Kondo effect is expected to manifest when the  $Pr^{3+}$  ions are diluted with elements without 4f electrons [2]. Bearing this in mind, we have prepared single crystalline samples of the diluted Pr system Y(Pr)Ir<sub>2</sub>Zn<sub>20</sub> [9]. The measurements of the specific heat and  $\rho$  showed that  $C_m/T$  follows  $-\ln T$  and  $\Delta \rho \propto \sqrt{T}$ , as predicted by the two-channel Kondo model [2]. Moreover, both data of  $C_m$  and  $\Delta \rho$  per Pr ion are well scaled as a function of  $T/T_0$ , where  $T_0$  is a characteristic temperature of the two-channel Kondo effect, as noted above. Therefore, we infer that the NFL behaviors result from the single-site quadrupole Kondo effect due to the hybridization of the  $4f^2$  state with two-channel conduction electrons.

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# Highly anisotropic strain dependencies in PrIr<sub>2</sub>Zn<sub>20</sub>

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We report thermal expansion and magnetostriction of the quadrupolar Kondo lattice  $PrI_2Zn_{20}$ , which exhibits antiferroquadrupolar order below  $T_Q = 0.11$ K and signatures of orbital Kondo Effect[1,2]. Linear thermal expansion and magnetostriction, measured along and perpendicular to magnetic fields B/[001], show a highly anisotropic tetragonal distortion which reflects the fundamental symmetry of the non-Kramers  $\Gamma_3$  ground state doublet, formed by the cubic  $T_d$  point group of the Pr-ions. Remarkably, the resulting volume change is vanishingly small, indicating very little variation of the Pr valence even at the critical field of antiferroquadrupolar order at B=5T along the [001] direction. We reveal that the novel magnetic field-induced state, which displays anomalous enhancements of Seebeck coefficient and strong softening of elastic constants[3,4], is only accompanied by a little variation of hybridization strength. Those results are in sharp contrast to Ce- and Yb-based Kondo lattice materials which show large volume changes as a function of magnetic field and temperature.

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# Field and angle dependent magnetoresistance in Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> thin films

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In 5*d* electron systems represented by iridates, spin-orbit and Coulomb interactions are of approximately the same order of magnitude. It has been theoretically predicted that as a result, novel topological phases such as Weyl semimetal and strongly correlated topological insulator states may emerge [1]. We have focused on a pyrochlore iridate,  $Pr_2Ir_2O_7$ , and found several interesting physical properties [2-5]: it is metallic down to the lowest temperatures but behaves as a spin liquid [2,3]; it exhibits spontaneous Hall effect generated by spin chirality [4]; and it has a Fermi node formed by quadratic band touching of the doubly degenerate valence and conduction bands at the  $\Gamma$ point at the Fermi level [5]. Moreover, we successfully fabricated (111)-oriented pyrochlore  $Pr_2Ir_2O_7$  epitaxial thin films [6]. The spontaneous Hall effect in the thin films appears up to about 50 K which is higher than the bulk report, and it is likely that the Weyl semimetal phase exists in the  $Pr_2Ir_2O_7$  thin films. In this study, we investigated the spin structure of the  $Pr_2Ir_2O_7$  thin films by angle-dependent magnetoresistance (MR) measurements, and discuss the chiral magnetic effect in these thin films.

Epitaxial (111)-oriented  $Pr_2Ir_2O_7$  thin films were fabricated on yttria-stabilized zirconia (111) substrates by pulsed laser deposition. MR measurements were performed with the four-terminal method, where the current (*I*) flows along the [110] direction.

Figure 1 shows the magnetic field dependence of MR ratio measured at 2 K in the configurations where the magnetic field (*B*) is either perpendicular ( $B \perp I$ , B//[110]) or parallel (B//I, B//[110]) to the current. These two experimental configurations are crystallographically equivalent. Both MR curves show negative MR due to the 2-in-2-out spin configuration. In contrast, the negative MR is larger when the magnetic field is parallel to the current. In the presentation, we will analyze the difference in the MR curves, and discuss the origin of the effect, such as a chiral magnetic effect.



Figure 1: Magnetic field dependence of the MR ratio of a  $Pr_2Ir_2O_7$  thin film measured at 2 K in the  $B \perp I$  and B//I configurations. The current flows along the [110] direction, "down" and "up" denote the sweep directions of the magnetic field.

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# Search for Multipolar Instability in URu<sub>2</sub>Si<sub>2</sub> Studied by Ultrasonic Measurements under Pulsed Magnetic Field

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The elastic properties of URu<sub>2</sub>Si<sub>2</sub> in the high-magnetic field region above 40 T, over a wide temperature range from 1.5 to 120 K, were systematically investigated by means of high-frequency ultrasonic measurements under the pulsed magnetic fields up to 68 T with pulse duration of about 150 ms at the Dresden High Magnetic Field Laboratory (HLD). The present study is motivated by a expectation that temperature dependence of the symmetry-breaking ultrasonic modes provides information on the bare 5*f*-electrons character with a broken of the unidentified electronic phase transition, so called 'hidden order', and weakened hybridization [1, 2]. We found contrast behaviors that appeared in three different transverse modes (as shown in Figs. 1); both  $\Gamma_4$  (B<sub>2g</sub>) and  $\Gamma_5$  (E<sub>g</sub>) symmetry modes of  $C_{66}$  and  $C_{44}$  show elastic softening which is enhanced above 30 T, respectively, while the characteristic softening of the  $\Gamma_3$  (B<sub>1g</sub>) symmetry mode ( $C_{11}-C_{12}$ )/2 is suppressed in high magnetic fields. These results underscore the presence of a hybridization-driven  $\Gamma_3$  (B<sub>1g</sub>) lattice instability [3] in the HO phase, and in

the strong *c*-*f* hybridization region at low-magnetic fields in URu<sub>2</sub>Si<sub>2</sub>. However, the enhanced softening of  $C_{44}$  and  $C_{66}$  for  $H \parallel [001]$  at high magnetic fields cannot be fully explained by using existing field crystalline-electric (CEF) schemes applied to the quadrupolar susceptibility in a local 5f<sup>2</sup> (J = 4) configuration, for example;  $(\Gamma_1^{(1)} - \Gamma_2)$  $-\Gamma_{3}$ ), or  $(\Gamma_{5}^{(1)} - \Gamma_{1}^{(1)} - \Gamma_{2})$ , or  $(\Gamma_{1}^{(5)} - \Gamma_{1}^{(5)} - \Gamma_{2})$  $\Gamma_{5}^{(2)} - \Gamma_{2}$ ), or  $(\Gamma_{3} - \Gamma_{1}^{(1)} - \Gamma_{2})$ , and so on. Thus, the origin of the softening in these transverse modes still remains an open question. Instead, we analyzed the softening of  $(C_{11}-C_{12})/2$  in lowmagnetic field region by using the phenomenological theory for the band-Jahn-Teller effect.

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Fig. 1. Magnetic-field and temperature dependence of the elastic constants (a,b,c):  $(C_{11}-C_{12})/2$ , (d,e,f):  $C_{66}$ , and (g,h,i):  $C_{44}$  for  $H \parallel [001]$  of URu<sub>2</sub>Si<sub>2</sub> at selected temperatures and magnetic fields.

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# Electronic structures suitable for the orbital Kondo effect and actual materials

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The orbital Kondo effect in the *f*-electron system without Kramers degeneracy is an interest research subject in the long term. However, there are little studies focused on the relationship between its origin and the electronic structure. In this work, firstly, we clarify necessary conditions of the occurrence about the orbital Kondo effect with the simplified model. As shown in Fig 1(left), we consider the band model in which the degeneration at  $\Gamma$  point and two conduction bands across Fermi level. Also at  $\Gamma$  point, the band structure has  $\Gamma_3$  symmetry. Considering freedom of the spin,  $\Gamma_3$  splits two states,  $\Gamma_3 = \Gamma_7 + \Gamma_8$ . In this time, *c-f* hybridization determines the condition of the Orbital Kondo effect. We propose candidates which we can observe orbital Kondo effect using the density of states and the hybridization from results of ab-initio calculation.

In this study, we report the result of PrPb<sub>3</sub>. In PrPb<sub>3</sub>, the band structure near Fermi level constituted the hybridized Pr-4*f* and Pb-6*p* electrons(Fig.1(middle)). In LaPb<sub>3</sub>, the conduction band has  $\Gamma_8^+$  symmetry at about 0.1Ry. above the Fermi level. In addition to the analysis of this electronic structure, we will discuss for the estimation of the orbital Kondo Temperature T<sub>K</sub>.



Fig. 1(left). Two degenerated conduction band at  $\Gamma$  point. (middle) electronic structure of PrPb<sub>3</sub> (right) electronic structure of LaPb<sub>3</sub>

# Observation of spin-split Fermi surfaces in the spin-orbit coupled metal Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub>

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Novel quantum phases emerging in the presence of strong spin-orbit coupling (SOC) are recently attracting particular attention in condensed matter physics. The multipolar phase with spontaneously broken inversion symmetry is one of the exotic phases predicted to be realized in correlated metals with SOC [1]. The lack of inversion symmetry lifts spin degeneracy and generates spin-polarized Fermi surfaces in a spin-orbit coupled metal. A promising candidate for hosting the SOC driven multipolar phase is the metallic 5*d* pyrochlore oxide Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub>. This compound undergoes two structural transitions at  $T_{s1} \sim 200$  K, and  $T_{s2} \sim 120$  K and the inversion symmetry is lost below  $T_{s1}$  [2]. Although the structural change across the transition at  $T_{s1}$  is extremely small, resistivity decrease steeply and the magnetic susceptibility shows a remarkable reduction. Therefore, the structural transition at  $T_{s1}$  is considered to originate from electronic instability. Recent optical second harmonic generation study has suggested the presence of a multipolar order below  $T_{s1}$  [3]. However, experimental evidence for spin-split Fermi surfaces expected for the multipolar order remains missing.

We present a quantum oscillation study of the spin-orbit coupled metal  $Cd_2Re_2O_7$ , which reveals spin-split Fermi surfaces of this compound for the first time. In the previous studies, it was difficult to detect quantum oscillation owing to the lack of high-quality single crystals. Our continuous effort to improve the sample quality has resulted in obtaining high-quality single crystals with residual resistivity ratio up to 300, which is one order of magnitude higher than those of previously prepared crystals. The high-quality crystals have enabled us to observe de Haas-van Alphen (dHvA) oscillations, clearly as shown in Fig. 1a. The fast Fourier transform spectrum of the dHvA oscillations (Fig. 1b) gives some dHvA frequency branches, which are proportional to the extremal cross-sectional areas of Fermi surfaces. We measured the angular dependence of the dHvA frequencies and observed spin-split electronic Fermi surfaces centered at the  $\Gamma$  point.

Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub> provides a unique opportunity to search for novel magnetoelectric effects realized in the multipolar ordered metals.



Fig. 1. (a) Field dependence of the magnetic torque at 30 mK with a magnetic field applied along the [111] direction. (b) Fast Fourier transform spectrum of the de Haas-van Alphen oscillation between 5 and 17.5 T.

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# Magnetic octupole induced large magneto-optical Kerr effect in the antiferromagnetic Weyl metal Mn<sub>3</sub>Sn

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When a polarized light beam is incident upon the surface of a magnetic material, the reflected light undergoes a polarization rotation. This magneto-optical Kerr effect (MOKE) has been intensively studied in a variety of ferro- and ferrimagnetic materials because it provides a powerful probe for electronic and magnetic properties as well as for various applications including magneto-optical recording. Recently, there has been a surge of interest in antiferromagnets (AFMs) as prospective spintronic materials for high-density and ultrafast memory devices, owing to their vanishingly small stray field and orders of magnitude faster spin dynamics compared to their ferromagnetic counterparts [1].

In fact, the MOKE has proven useful for the study and application of the antiferromagnetic (AF) state. Although limited to insulators, certain types of AFMs are known to exhibit a large MOKE, as they are weak ferromagnets due to canting of the otherwise collinear spin structure [2]. In the fully compensated collinear AFMs, where the MOKE is usually absent, quadratic magneto-optical effects such as the Voigt effect can be useful to determine the Néel vector [3]. On the other hand, recent theoretical and experimental progress has revealed that systems such as certain spin liquids and non-collinear AFMs can exhibit a large anomalous Hall effect (AHE) in zero applied magnetic field despite a vanishing magnetization [4-8]. Because the AHE has the same symmetry requirements as the MOKE, it is possible that the same class of AFMs may exhibit a Kerr rotation. Thus, the recent experimental discovery of a large AHE in the non-collinear AFM Mn<sub>3</sub>Sn [7], which is recently reported to be a Weyl magnet [9], as well as its soft response to a magnetic field give promise for a potentially large MOKE character.

Here we report the first observation of a large MOKE signal in an AF metal at room temperature [10]. In particular, we find that despite a vanishingly small magnetization of  $M \sim 0.002$   $\mu_B/Mn$ , the non-collinear AF metal Mn<sub>3</sub>Sn exhibits a large zero-field MOKE with a polar Kerr rotation angle of 20 milli-degrees, comparable to ferromagnetic metals. Our first-principles calculations have clarified that ferroic ordering of magnetic octupoles in the non-collinear Néel state may cause a large MOKE even in its fully compensated AF state without spin magnetization. This large MOKE further allows imaging of the magnetic octupole domains and their reversal induced by magnetic field. The observation of a large MOKE in an AF metal shall open new avenues for the study of domain dynamics as well as spintronics using AFMs.

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# Theory of the flux-flow Hall effect in an isolated vortex of an s-wave superconductor

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After the sign change of the Hall conductivity have been observed in some cuprate superconductors, intensive studies have been performed on the flux-flow Hall effect in type-II superconductors theoretically and experimentally. Despite these efforts, a microscopic understanding of the anomalous flux-flow Hall effect is still missing. This may be because the Lorentz force is missing from the standard Eilenberger equations, which have been used extensively to study vortices quantitatively and are now regarded as a basic and reliable tool for investigating inhomogeneous and/or nonequilibrium superconductors microscopically. In fact, the standard equations cannot describe the vortex-core charging and flux-flow Hall effect.

Recently, the Lorentz force has been incorporated successfully in a gauge-invariant manner within the real-time Keldysh formalism [1]. The augmented quasiclassical equations in the Keldysh formalism have been used to study charging in the Meissner state with Fermi surface and gap anisotropies [2], and also to calculate flux-flow Hall conductivity numerically for the s-wave pairing on an isotropic Fermi surface [3]. However, the temperature dependence of the Hall conductivity has not been calculated in Ref. [3].

On the other hand, we derived augmented quasiclassical equations of superconductivity with the Lorentz force in the Matsubara formalism so that the charge redistribution due to supercurrent can be calculated quantitatively [4]. It is still desirable when studying the charging to transform the equations into the Matsubara formalism, in which equilibrium properties and linear responses can be calculated much more easily.

We develop a new approach to calculate the liner responses in the flux-flow state by transforming the energy variable of the augmented quasiclassical equations in the Keldysh formalism into the Matsubara energy on the imaginary axis. It is shown that linear responses can be calculated much more easily compared to the approach based on the augmented quasiclassical equations in the Keldysh formalism. Using it, we confirm that there exists the ohmic and Hall resistivity caused by the moving isolated vortex in an s-wave superconductor, and also calculate the temperature dependence of the Hall angle in flux-flow state.

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# High pressure studies of quadrupole order and superconductivity in PrT<sub>2</sub>Al<sub>20</sub> (T=Ti, V)

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 $PrT_2Al_{20}$  (T=Ti, V) with non-Kramers  $\Gamma_3$  doublet ground state in the cubic crystal structure provides the unique opportunity to study the interplay between the quadrupole order and superconductivity [1,2]. In particular, PrTi<sub>2</sub>Al<sub>20</sub> exhibits the enhancement of superconducting transition temperature near the putative quantum critical point of the ferroquadrupole order [3], suggesting that quadrupole fluctuations may provide a nonmagnetic glue for Cooper pairing. More interestingly, the strong hybridization between the ground doublet and conduction electrons gives rise to the quadrupolar Kondo effect, leading to an anomalous metallic state. Indeed, the non-Fermi liquid (NFL) behavior, possibly associated with the quadrupole Kondo effect, has been observed in antiferroquadrupolar PrV<sub>2</sub>Al<sub>20</sub> with strong hybridization effect [2,4]. However, the underlying mechanisms for the appearance of superconductivity and the associated fluctuations remain unclear. Here we present the effect of pressure on PrT<sub>2</sub>Al<sub>20</sub> (T=Ti, V) to shed light on the interplay between the quadrupole order and superconductivity. In both compounds, applying pressure suppresses the quadrupole order and a dome-like superconducting phase emerges, accompanied by the strong enhancement of the low temperature resistivity caused by the stronger hybridization. We also found that the NFL state emerges in wide pressure and field range, where the resistivity curves are shown to collapse onto a single scaling function, as predicted for the quadrupole Kondo lattice model [5]. In this presentation, we discuss a generic feature in Pr1-2-20 compounds under strong hybridization on the bases of the temperature-pressure-field phase diagram.

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# Anomalous Hall effect and magnetoresistance in the nodal metallic spin ice Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>

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Pyrochlore  $Pr_2Ir_2O_7$  is a rare material with various unique properties. Unlike the other pyrochlore iridates,  $Pr_2Ir_2O_7$  does not exhibit metal-insulator transition and keeps metallic behavior without long-range magnetic order[1]. Magnetic Grüneisen ratio diverges  $\Gamma_{mag} \sim T^{-3/2}$  without tuning any parameter, indicating the zero-field quantum criticality [2]. Besides, recent angle-resolved photoemission spectroscopy (ARPES) measurement reveals the Fermi node at  $\Gamma$  point in Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, which can be an origin of the various topological phases such as topological insulator and Weyl semimetal [3].

One of the most interesting properties of  $Pr_2Ir_2O_7$  is unusual anomalous Hall effect; spontaneous Hall effect appears even in the absence of any spin freezing, which is attributed to the chiral spin liquid state [4]. In this poster, we will discuss the recent results for the anomalous Hall effect for various samples of  $Pr_2Ir_2O_7$ .

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## Time-Dependent Reentrant Superconductivity in Nonequilibrium KBi<sub>2</sub>

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Nonequilibrium states or hidden states of materials, such as photo-induced metallic state, are attracting considerable interests. These nonequilibrium states can be induced by irradiating laser pulses or decreasing film thickness, for instance [1,2]. In this presentation, we will report a novel nonequilibrium states induced by a specific thermal process in a Laves phase superconductor KBi<sub>2</sub>. The compound KBi<sub>2</sub> has been reported to exhibit superconductivity below a transition temperature  $T_c = 3.6$  K and crystallize in a cubic MgCu<sub>2</sub>-type structure (space group Fd $\overline{3}$ m, O<sub>h</sub><sup>7</sup>, #227) [3,4].

We observed bulk superconductivity at  $T_c = 3.6$  K in KBi<sub>2</sub> when the synthesis procedure in Ref. 4 was used. On the other hand, superconductivity disappeared (or  $T_c$  became below 1.9 K), when a specific heating process (rapid heating and cooling) was applied, as shown in Fig. 1(a). Interestingly, we observed that bulk superconductivity recovered when the sample was stored at room temperature for a long enough time (~500 hours) following another thermal cycle between room temperature and low temperature (1.8 K), as shown in Fig. 1(e). In the middle of recovery process, reentrant superconductivity appeared as shown Fig. 1(b,c).

Several materials such as URhGe and  $\kappa$ -(BETS)<sub>2</sub>FeBr<sub>4</sub> are known to exhibit reentrant superconductivity, because of the interplay between superconductivity and magnetic ordering [5,6]. However, KBi<sub>2</sub> consists of no magnetic elements, thus a novel mechanism should be invoked.



Fig. 1. Magnetization divided by field M/H as a function of temperature in KBi<sub>2</sub> for various storing times t at room temperature. The measurements were performed in a zero-field-cooling condition with an applied magnetic field H = 10 Oe

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# Low Temperature Synthesis and Specific Heat of Chevrel Phase Compounds Mo<sub>6</sub>Ch<sub>8</sub> (Ch = S, Se, Te)

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Chevrel compounds Mo<sub>6</sub>S<sub>8</sub>, Mo<sub>6</sub>Se<sub>8</sub>, and Mo<sub>6</sub>Te<sub>8</sub> crystalize in a rhombohedral structure with the space group  $R\overline{3}$  ( $C_{3i}^2$ , No. 148). These isostructural and isoelectronic compounds exhibit quite different superconducting transition temperatures  $T_c$ , namely  $T_c = 1.8$  and 6.3 K for Mo<sub>6</sub>S<sub>8</sub> [1] and Mo<sub>6</sub>Se<sub>8</sub> [2], respectively, while Mo<sub>6</sub>Te<sub>8</sub> does not exhibit superconductivity [3]. In order to investigate the reason why the  $T_c$  of Mo<sub>6</sub>Se<sub>8</sub> is so high compared with other two compounds, we performed specific heat measurements using high quality polycrystalline samples obtained by a low temperature synthesis.

The polycrystalline samples of  $Mo_6S_8$ ,  $Mo_6S_8$ , and  $Mo_6Te_8$  were synthesized in two steps. First, polycrystalline  $Cu_2Mo_6Ch_8$  (Ch = S, Se, Te) were synthesized by a solid-state reaction. Then, Cu was removed by leaching in a HCl/H<sub>2</sub>O solution. The obtained samples were characterized by powder X-ray diffraction. When  $Cu_2Mo_6Ch_8$  was synthesized, the addition of excess Mo was found to be useful to suppress the formation of impurity phase MoCh<sub>2</sub> (Ch = S, Se, Te). The remaining excess Mo was removed completely after dissolved in the HCl/H<sub>2</sub>O solution.

Specific heat measurements were carried out using PPMS (Quantum Design). The temperature dependence of specific heat for  $Mo_6Se_8$  exhibited a clear jump at  $T_c = 6.2$  K, indicating the emergence of bulk superconductivity. On the other hand, the specific heat of  $Mo_6S_8$  and  $Mo_6Te_8$  did not show superconducting behavior above 2.0 K. The analysis of normal-state specific heat data revealed that the high  $T_c$  of  $Mo_6Se_8$  originated from the large electronic density of states at the Fermi level and soft-phonons, thus, a strong electron-phonon coupling.

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# Superconductivity in Mg<sub>2</sub>Ir<sub>3</sub>Si: An Ordered Variant of the Hexagonal Laves Phase MgZn<sub>2</sub>

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Laves phase is the intermetallic compounds which are described as AB<sub>2</sub>. They crystallize in either a cubic MgCu<sub>2</sub> type, a hexagonal MgZn<sub>2</sub> type, or a hexagonal MgNi<sub>2</sub> type structure. There are many superconductors in the Laves phase. Among them, the hexagonal MgZn<sub>2</sub>-type ScTc<sub>2</sub> exhibits the highest  $T_c$  of 10.9 K [1]. However, radioactive tecnetium prohibited us from further studying the origin of high  $T_c$  in ScTc<sub>2</sub>. This motivated us to develop a novel hexagonal MgZn<sub>2</sub>-type superconductor with high  $T_c$ . In this study, we report the discovery of novel compound Mg2Ir<sub>3</sub>Si, which is an ordered variant of the hexagonal Laves phase MgZn<sub>2</sub>. The compound exhibited superconductivity at 7 K. This is the highest value of  $T_c$  among the hexagonal MgZn<sub>2</sub>-type phase.

Polycrystalline samples of  $Mg_2Ir_3Si$  were synthesized by a solid state reaction. The structures of obtained samples were examined by powder X-ray diffraction (XRD). Rietveld refinement was performed using the RIETAN-FP program [2]. The magnetization was measured using MPMS (Quantum Design). The electrical resistivity and the specific heat were measured using PPMS (Quantum Design).

The powder XRD profiles of the obtained samples were refined based on the hexagonal Mg<sub>2</sub>Cu<sub>3</sub>Si-type structure with the space group  $P6_3/mmc$  ( $D_{6h}^4$ , No. 194). The type of structure corresponds to the ordered variant of MgZn<sub>2</sub>. The Ir and Si atoms of Mg<sub>2</sub>Ir<sub>3</sub>Si occupy two different crystallographic sites of the Zn in MgZn<sub>2</sub>. The temperature dependences of magnetization, electrical resistivity, and specific heat revealed that the Mg<sub>2</sub>Ir<sub>3</sub>Si was a bulk superconductor with  $T_c = 7$  K. The comparison of the electronic specific heat coefficient between the present material and the MgZn<sub>2</sub>-type low temperature superconductors [3] showed that the high  $T_c$  of Mg<sub>2</sub>Ir<sub>3</sub>Si originated from the high electronic density of states (DOS) at the Fermi level. The first-principles calculations suggested that the DOS at the Fermi level was dominated by Ir 5*d*. This warrants the importance of further studies because the locally noncentrosymmetric structure of Ir layers and a strong spin-orbit coupling of Ir are a prerequisite for exotic superconductivity.

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# Effect of elemental substitution for SnAs-based novel layered superconductor NaSn<sub>2</sub>As<sub>2</sub>

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Superconductivity with exotic properties has often been discovered in materials with a layered (two-dimensional) crystal structure. The low dimensionality affects the electronic structure of materials, which could realize a high transition temperature ( $T_c$ ) and/or unconventional pairing mechanisms. Here, we report the superconductivity in a layered tin arsenide NaSn<sub>2</sub>As<sub>2</sub> [1]. As shown in Fig. 1(a), the crystal structure consists of  $(Sn_2As_2)^{2-}$  bilayers, which is bound by van-der-Waals forces, separated by Na<sup>+</sup> ions [2]. The  $(Sn_2As_2)^{2-}$  conducting layers have the buckled honeycomb-type structure (Fig. 1(b)). Measurements of electrical resistivity and specific heat confirm the bulk nature of superconductivity of NaSn<sub>2</sub>As<sub>2</sub> with  $T_c$  of 1.3 K. Our results propose that the SnAs layers will be a basic structure providing another universality class of a layered superconduction family, and it provides a new platform for the physics and chemistry of low-dimensional superconductors. In the conference, we also present the effect of the elemental substitution for NaSn<sub>2</sub>As<sub>2</sub>, including the synthesis of the novel materials.



Figure 1. (a) and (b) Crystal structure of NaSn<sub>2</sub>As<sub>2</sub>. (c) Temperature dependence of electrical resistivity of NaSn<sub>2</sub>As<sub>2</sub>.

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# Temperature and Field Dependence of Lattice Elasticity in Quantum Spin Ice Material Pr<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub>

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Geometrical frustration suppresses a long-range order and gives rise to macroscopically degenerate ground states. Pyrochlore oxides, for example, Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, are typical frustrated magnets whose ground state has 2-in 2-out configuration and known to be classical spin ice [1]. On the other hand, quantum fluctuation can play an important role, when the magnetic element is replaced with the one with a smaller spin such as Pr, which lifts the macroscopic degeneracy of the spin ice. Pr<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> and Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> are the candidates of such *quantum* spin ice materials. Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> is metallic and exhibits anomalous Hall effect without magnetic field or magnetization, indicating the possible chiral spin liquid [2]. Compared to Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub>, *insulating* Pr<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> allows us to study much simpler quantum spin ice physics due to the absence of the conduction electrons. Pinch points, which are indications of spin-ice correlation are observed in Pr<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> by the neutron scattering experiment [3]. In addition, an anomaly is reported from the recent thermal conductivity measurement [4]. Thermal conductivity increases below 200 mK and shows a peak around 80 mK. Since there are no thermal carriers at such low temperatures, the theoretically proposed new quasiparticle "photon" must be the carriers [5]. Pr<sup>3+</sup> is a non-Kramers ion, and sensitively reacts to the lattice strain hence thermal expansion and magnetostriction become crucial. By application of the magnetic field along Ising axis B // [111], classical spin ice materials show a first-order, liquid-gas type transition [6]. On the other hand, such a transition for quantum spin ice has never been well studied experimentally. Thus, we performed the magnetization, magnetostriction and thermal expansion measurements at ultra-low temperatures. In this poster session, I will report the result of these measurements, using high quality single crystal of Pr<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub>.

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# Se Isotope Effects of LaO<sub>0.6</sub>F<sub>0.4</sub>Bi(S,Se)<sub>2</sub> Superconductor

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Layered superconductors have drawn much attention owing to the observations of high-transition-temperature (high- $T_c$ ) superconductivity and unconventional pairing mechanisms in cuprate- and Fe-based superconductors. The layered bismuth chalcogenide (BiCh<sub>2</sub>-based) superconductor [1], discovered in 2012, is a new class of layered superconductor with a maximum  $T_c$  of 11K, but the pairing mechanisms of the superconductivity in the BiCh<sub>2</sub>-based system have not been understood. Although early theoretical calculations [2] and experiments [3] proposed conventional phonon-mediated pairing mechanism, resent theoretical calculations [4] and angle-resolved photoemission spectroscopy (ARPES)[5] proposed unconventional pairing mechanisms for the superconductivity of BiCh<sub>2</sub>-based system.

Here, we report the Se isotope effect on BiCh<sub>2</sub>-based superconductor  $LaO_{0.6}F_{0.4}Bi(S,Se)_2$  with <sup>76</sup>Se and <sup>80</sup>Se isotopes. Because superconductivity emerges in the BiCh-plane, and the conduction band is composed of Bi-6p orbitals hybridized with Ch-p orbitals, the Se isotope effect should be an useful probe to examine the importance of phonon for superconductivity of this compound.

Measurement results of magnetization and resistivity showed that  $T_c$  did not change between the <sup>76</sup>Se and <sup>80</sup>Se samples. This fact indicates that the exponent  $\alpha_{Se}$  is close to zero, while BCS theory gives  $\alpha \sim 0.5$ . Our results suggest that pairing in the BiCh<sub>2</sub>-based superconductors is not mediated by phonons, and unconventional superconductivity may emerge in the BiCh<sub>2</sub> layer [6].



Fig. 1. Crystal structure of LaO<sub>0.6</sub>F<sub>0.4</sub>Bi(S,Se)<sub>2</sub>.

Fig. 2. Temperature dependence of magnetization for the LaO0.6F0.4Bi(S,Se)2 with  $^{76}$ Se and  $^{80}$ Se.

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# Synthesis of bismuth-chalcogenide layered superconductor with Ce-based blocking layer

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Since the discovery of BiCh<sub>2</sub>-based (Ch: S, Se) superconductors in 2012, this family of compounds has received much attention as a new class of layered superconductors. The crystal structure composed of alternate stacks of electrically conducting BiCh<sub>2</sub> layers and blocking layers is similar to that of the cuprate or Fe-based high-transition-temperature (high-T<sub>c</sub>) superconductors. [1]

Regarding carrier doping method for REOBiCh<sub>2</sub>-type compounds (RE: rare earth), partial substitutions of F<sup>-</sup> for O<sup>2-</sup> ions have been typically employed to induce superconductivity. In the present study, we report the effects of Ce substitution for the RE site on structural, electronic, and magnetic properties in La<sub>1-x</sub>Ce<sub>x</sub>OBiSSe (x = 0-0.9), which are newly obtained in this study. Undoped (x = 0) sample showed semiconducting-like transport below 100 K, metallic conductivity was observed for  $x \ge 0.1$ . This is because electron carriers are induced by mixed valence of Ce ions, as revealed by bond valence sum calculation and Curie-Weiss analysis of magnetization measurements.

Zero resistivity and clear diamagnetic susceptibility were obtained for x = 0.2-0.6, implying the emergence of bulk superconductivity in these chemical compositions. Dome-shaped superconductivity phase diagram with the highest transition temperature of 3.1 K for x = 0.3-0.5 was established, as shown in Fig. 1.

The present study clearly shows the emergence of bulk superconductivity and metallic conductivity via mixed valence of Ce ions in layered bismuth-chalcogenide systems [2].



Fig. 1 Superconductivity phase diagram of La<sub>1-x</sub>Ce<sub>x</sub>OBiSSe.

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# Hall effect in Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub> under high pressure

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Pyrochlore superconductor Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub> shows two structural transitions at ambient pressure. Cubic pyrochlore structure at room temperature (phase I) transforms to the tetragonal structure without inversion symmetry in phases II and III. The transition at  $T_{s1}$  has been regarded as a band Jahn-Teller transition due to the band structure with high degeneracy in the high-symmetric pyrochlore structure. As a result, tiny change of crystal deformation cases the drastic change in the transport properties. [1] Application of pressure suppresses these transition temperatures and induces many phases (IV-VIII). [2] The superconducting transition temperature  $T_c$  increases with increasing pressure in phase III and reveals no drastic change in the pressure-induced phases IV, VII and VIII. On the other hand, the upper critical field  $B_{c2}$  increases drastically with increasing pressure. Recently the theoretical scenario was reported, where the fluctuation of  $T_{s1}$  transition generate an attractive pairing interaction in an odd-parity pairing channel, which promotes the odd-parity superconducting state. [3]

In this study, we carried out the Hall effect measurement under high pressure in order to investigate the variation of the electronic state. As shown in Fig. 1, the temperature dependence of Hall coefficient  $R_{\rm H}$  at ambient pressure agree with the previous result [1].  $R_{\rm H}$  shows the positive value in phase I and changes the sign in phase II. The absolute value  $|R_{\rm H}|$  increases in phase III. The decrease of  $|R_{\rm H}|$  with increasing pressure is considered to relate to the increase of  $T_{\rm c}$  and  $B_{\rm c2}$ . Further increase of  $|R_{\rm H}|$  is observed in the pressure-induced phase IV. In Fig. 1,  $R_{\rm H}$  is determined near zero field. The field dependence of Hall resistance shows a straight line in phases I and II but is curved in phases III and IV. The linear field dependence is reproduced by assuming the two carrier system in the case of the compensated metal while the curved that indicates the three carrier model.



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

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# Field-orientation dependence of a ferroquadrupole order in PrTi<sub>2</sub>Al<sub>20</sub> investigated by thermal measurements

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The cubic compound  $PrTi_2Al_{20}$  has the ground state of the non-Kramers  $\Gamma_3$  doublet under the crystalline electric field (CEF) and exhibits a multipole order at  $T_Q \sim 2$  K [1]. It is attributed to a ferroquadrupole order because of a broadening of the transition under a magnetic field [1], and moreover, the absence of a magnetic superlattice reflection in the neutron scattering experiment in a magnetic field [2]. Recently, NMR measurements [3] have revealed the occurrence of a field-induced 1st-order transition at roughly 2 T when the magnetic field is applied along the [100] and [110] axes, whereas it is absent in the [111] direction. This anisotropic phase diagram suggests a variety of quadrupole ordered states in  $PrTi_2Al_{20}$ , which is key to understand the physics of the multipolar system. On the basis of a phenomenological model with mean-field approximation [4], this phase transition is explained by competition among the CEF effect, interaction between quadrupoles, and the non-linear Zeeman effect on quadrupole moments. Furthermore, it is proposed that the model can reproduce the anisotropic phase diagram more accurately if the interaction between quadrupoles is assumed to depend on the magnetic field [3].

In order to obtain further information on this anisotropic phase diagram, we measured the specific heat of a high-quality single crystalline  $PrTi_2Al_{20}$  under a rotating magnetic field (up to 5 T) within the [1-10] plane. In this study, a magnetic-field orientation was controlled threedimensionally by using a vector magnet, generating the magnetic field within the *xz* plane, and using a stepper motor that is mounted on the top of the Dewar to rotate a refrigerator around the *z* axis. Then, we found that the transition at  $T_Q \sim 2$  K is nearly independent of *H* and remains 2nd order in the [111] direction, whereas it becomes broad and crossover with increasing *H* in [001] and [110] directions. In addition, we revealed that the broadening of the transition in *H* // [110] is limited to the high-temperature region near  $T_Q$ ; the low-temperature specific heat is insensitive to *H* in the [110] direction, whereas it significantly changes in *H* // [001]. This fact indicates that the Zeeman effect hardly changes the energy gap between the ground and excited states in *H* // [110], at least below 5 T. From field-rotation measurements, we uncovered that the 2nd-order transition is rigid only when the magnetic field is applied precisely along the [111] direction. These experimental facts support the recently-proposed model that assumes the field-dependent interaction between quadrupolars, and deepen the understanding of multipole nature in this system.

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# Superconductivity in IrIn<sub>2</sub> with Iridium Infinite Chains: A Comparative Study of CoIn<sub>2</sub> and IrIn<sub>2</sub>

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IrIn<sub>2</sub> and CoIn<sub>2</sub> crystallize in the orthorhombic Mg<sub>2</sub>Cu-type structure (space group, *Fddd*,  $D_{2h}^{24}$ , No.70) [1]. In this research, we discovered superconductivity in IrIn<sub>2</sub> at a superconducting transition temperature  $T_c = 2.2$  K. The isostructural and isoelectronic compound CoIn<sub>2</sub> did not exhibit superconductivity down to 1.8 K.

IrIn<sub>2</sub> was synthesized by heating a mixture of Ir and In. The mixture was heated at 1050 °C for 24 h. Then, it was quenched in cold water. CoIn<sub>2</sub> was synthesized by heating a mixture of Co and In. The mixture was heated at 170 °C for 5 h and then at 500 °C for 200 h. Then, it was quenched in cold water. RhIn<sub>2</sub> phase could not be synthesized in consistent with the Rh-In binary equilibrium phase diagram.

The magnetization and resistivity revealed that  $IrIn_2$  is a superconductor with  $T_c = 2.2$  K, as shown in Figs. 1(a) and 1(b). No superconductivity was observed in CoIn<sub>2</sub>. The specific heat of  $IrIn_2$  exhibited a superconducting transition at ~ 2.2 K, as shown in Fig. 1(c). We estimated the electronic specific-heat coefficient  $\gamma = 6.3$  and 13.8 mJ/K<sup>2</sup>·mol and the Debye temperature  $\theta_D =$ 222 and 243 K for IrIn<sub>2</sub> and CoIn<sub>2</sub>, respectively. The absence of superconductivity in CoIn<sub>2</sub> is unusual because larger  $\gamma$  tends to exhibit higher  $T_c$  in the most family of superconductors. Firstprinciples calculations for CoIn<sub>2</sub> showed the presence of a peak in DOS at around the Fermi level  $E_F$ . This suggests that CoIn<sub>2</sub> is located near ferromagnetic instability, and thus superconductivity is suppressed. The calculation showed that Ir 5d orbital dominates the DOS at  $E_F$ , thus, superconductivity in IrIn<sub>2</sub>.



Figure 1. The temperature dependence of (a) magnetization M and (b) electrical resistivity  $\rho$  for IrIn<sub>2</sub>, and (c) C/T versus  $T^2$  plot for IrIn<sub>2</sub> and CoIn<sub>2</sub>. The solid lines denote a fit by  $C/T = \gamma + \beta T^2$ .

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# Intrinsic Origin Investigation of AHE and Thermoelectric in Weyl Semimetal Mn<sub>3</sub>Ge

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Intrinsic (Berry curvature) origin of the anomalous Hall effect (AHE) once had been a controversy for decades in ferromagnetic systems. Since there are three mechanisms account for the AHE: skew-scattering, side-jump and Berry curvature, which dominates in the super clean and good magnetic metals, respectively [1]. Especially, in ferromagnets it is very difficult to differentiate the AHE contributions between side-jump and Berry curvature mechanism, because they have the same longitudinal resistivity dependence,  $\rho_{AH} \propto \rho^2 M$ , where  $\rho$  and M are longitudinal resistivity and magnetization.

Recently, the observation of the AHE in antiferromagnets has been reported for the inverse triangle spin texture  $Mn_3Sn$  [2-4] and  $Mn_3Ge$  [5,6]. Different from the ferromagnets, this coplanar but noncollinear spin structure does not induce magnetization or very tiny of about a few  $m\mu_B/Mn$  with inplane directions, which offer a very ideal platform for the investigation on intrinsic (Berry Curvature) origin. Here, we show our recent work on the study of the AHE of  $Mn_3Ge$ .

Single crystals of Mn<sub>3</sub>Ge with size of about 1 cm length have been grown. Systematic transport measurement results show that the temperature cooling resistivity along [0001] and [2-1-10] directions has typical metallic behavior with the residual resistivity ratio (RRR) about 3. Interestingly, with increasing temperature from 2K to 200K the Hall resistivity can also be several times larger, while the magnetization changes very tiny, indicating the failure of the conventional AHE formula, and requires the Berry curvature description. Besides, Nernst signal was found to be rather large and evolve with temperature, which most likely comes from the Berry curvature at deferent chemical potential position.

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# Odd-parity multipoles by orbital hybridization in noncentrosymmetric tetragonal systems

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Electrons in solids have the multiple degrees of freedom, such as charge, spin, and orbital. Their interplay leads to various electronic phases and intriguing transport properties. Among them, the interplay between the spin and orbital degrees of freedom are described by multipoles in a unified manner, and it is useful to characterize the anisotropic electric charge and current distributions [1]. The multipole orderings were detected in *f*-electron systems, e.g., an electric quadrupole ordering in  $PrIr_2Zn_{20}$  [2] and a magnetic octupole ordering in  $Ce_{1-x}La_xB_6$  [3]. The studies for such multipole orderings have been mainly done for the centrosymmetric systems without the orbital hybridization. In that situation, the only even-parity multipoles are activated. Meanwhile, there is a chance to have an odd-parity multipole in noncentrosymmetric systems through the orbital hybridization with parity mixing [4]. Furthermore, the recent study shows that a 'toroidal multipole', which is distinguished from conventional electric and magnetic multipoles, can be activated by the orbital hybridization [5]. As a next step, it is intriguing to explore what odd-parity multipoles including toroidal multipoles are activated in specific crystal structures.

In the present study, we examine how the odd-parity multipoles are induced on specific lattice structures. We here consider a possible multipole through the orbital hybridization by taking three tetragonal systems (i)  $D_{4h}$  with the inversion symmetry, (ii)  $D_{2d}$  and (iii)  $C_{4v}$  without the inversion symmetry. For these systems, we investigate the role of the hybridization in inducing odd-parity multipoles and classify them according to each point group. For example, we consider two orbitals without the *d*-*f* hybridization in the point group  $D_{4h}$ :  $\phi_{xy}$  with the symmetry  $B_{2g}$  and  $\phi_{xyz}$  with the symmetry  $B_{1u}$  as shown in Fig. 1. These orbitals hybridize in the point group  $C_{4v}$ , since they belong to the same irreducible representation  $B_2$ . As a result, the toroidal dipole degree of freedom  $T_z$  is activated as the inter-orbital degree of freedom. In the presentation, we show what hybridizations activate the toroidal multipole degree of freedom for three tetragonal systems and discuss what multipole orderings are stabilized in the ground state within the mean-field calculations.



Fig. 1. The energy splitting of d- and f-orbital in the crystalline electric field with the D4h and C4v symmetry

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# The inelastic neutron scattering study on the strange metal behavior in the intermediate valence system α-YbAlB<sub>4</sub>

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Among a number of 4*f* electron systems,  $\beta$ -YbAlB<sub>4</sub> is particularly interesting as it exhibits the first type of heavy fermion superconductivity in Yb-based heavy fermion systems and unconventional quantum criticality at ambient pressure [1,2]. Moreover, recent studies have shown that the quantum criticality appears in a finite range of pressure and is separated from a magnetic instability under pressure by a Fermi liquid phase [3].

Its sister compound  $\alpha$ -YbAlB<sub>4</sub> has a Fermi liquid ground state [4]. However, it has the same local structure around Yb and exhibits strong valence fluctuation as same as  $\beta$ -YbAlB<sub>4</sub> [5]. From the theoretical aspect [6], its sevenfold symmetric environment of the Yb plays a key role on the hybridization and its quantum criticality. Therefore, the possibility of emergence of strange metal also in  $\alpha$ -YbAlB<sub>4</sub> is pointed out. Recently and actually, such behaviour is observed with applying external magnetic field 3.3 T with accompanying metamagnetism [7].

In order to investigate this strange metal behaviour from the microscopical view, we performed the inelastic neutron scattering measurement on  $\alpha$ -YbAlB<sub>4</sub>. We will discuss the origin of this strange metal behaviour from this result, especially focusing on whether it comes from magnetic instability or not.

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# Magnetic Properties of Hexagonal Yb<sub>1-x</sub>Lu<sub>x</sub>CuGe

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YbCuGe crystallizes in the hexagonal LiGaGe-type structure and orders antiferromagnetically below  $T_{\rm N} = 4.2$  K [1]. The Sommerfeld coefficient  $\gamma$  for YbCuGe shows small value  $\gamma = 5.2$  mJ/mol K<sup>2</sup>. The determined value of magnetic entropy  $S_{\rm m}$  at  $T_{\rm N}$  is 28 % of Rln2, expected for a doublet ground state. It is suggested that the slow release of magnetic entropy is probably ascribed to the spin fluctuations due to the geometrical frustration effects.

In order to obtain more insight into the nature of the geometrical frustration effects in this system, we studied single crystals of  $Yb_{1-x}Lu_xCuGe$  with x from 0 to 1. Single crystals of  $Yb_{1-x}Lu_xCuGe$  were grown by the Bridgman-method using a sealed tungsten crucible. Figure 1 shows the magnetic phase diagram and the Sommerfeld coefficient  $\gamma$  of  $Yb_{1-x}Lu_xCuGe$  as a function of Lu concentration x. Substitution of nonmagnetic Lu for magnetic Yb ions reduces  $T_N$  and the antiferromagnetic order vanishes around x = 0.6. We found that the  $\gamma$  increases with increasing x.



Fig. 1 Magnetic phase diagram and the Sommerfeld coefficient g of Yb<sub>1-x</sub>Lu<sub>x</sub>CuGe.

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# <sup>73</sup>Ge-NQR studies under pressure on magnetic fluctuations of ferromagnetic superconductor UGe<sub>2</sub>

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UGe<sub>2</sub> is a ferromagnet with the ferromagnetic (FM) Curie temperature  $T_{\text{Curie}} = 52$  K at ambient pressure and the  $T_{\text{Curie}}$  is suppressed to 0 K with increasing the pressure. The superconducting (SC) transition was observed in the pressure range of P = 1 to 1.5 GPa.[1] The magnetization shows the enhancement at  $T_X$  in the FM phase.[2] The transition between FM1 with smaller ordered moment and FM2 with larger one is a broad crossover at ambient pressure. However, the crossover region terminates by applying the pressure. The terminal point is called a critical point (CP), above which the FM1-FM2 transition is of a first order. The CP in UGe<sub>2</sub> was reported as  $T_{\text{CP}} = 7$  K and  $P_{\text{CP}} = 1.16$ GPa. The FM1-FM2 transition disappears above P = 1.2 GPa where the superconducting transition temperature is highest. This suggests the SC is involved in the FM1-FM2 transition in UGe<sub>2</sub>.

The NMR measurements were performed in UCoGe which is one of the ferromagnetic superconductors. The measurements of the spin-lattice relaxation rate  $1/T_1$  in UCoGe indicated that UCoGe possesses the Ising type magnetic fluctuation along the magnetic easy axis (*c*-axis).[3] In addition, it was reported that the anisotropic fluctuation correlates closely with the SC.[4] On the other hand, the details of the magnetic fluctuations in UGe<sub>2</sub> have not been reported. In order to clarify the anisotropy of the magnetic fluctuations in UGe<sub>2</sub>, we carried out the <sup>73</sup>Ge-NQR measurement under pressure.

We measured  $T_1$  and the spin-spin relaxation time  $T_2$  to clarify the anisotropy of the magnetic fluctuations under several pressures. Figure 1 shows the temperature dependences of  $1/T_1T$  and  $1/T_2$  at P = 0.71, 0.98, and 1.31 GPa. Below  $T_X$ , the suppression of  $1/T_1T$  is observed at 0.71 and 0.98 GPa. On the other hand,  $1/T_2$  is enhanced near  $T_X$ . The behaviour of  $1/T_2$  shows the large divergence at 0.98 GPa, where is close to  $P_{CP}$ . These results suggest that the magnetic fluctuations along *a* axis is enhanced toward the CP. The peak of  $1/T_2$  at 1.31 GPa, where the FM1-FM2 transition disappears, is observed at 3 K. We are now measuring  $T_1$  and  $T_2$  at 1.21 GPa to investigate the anomaly of  $1/T_2$  at 1.32 GPa.



Figure 1. Temperature dependences of  $1/T_1T$  and  $1/T_2$  at P = 0.71, 0.98 and 1.32 GPa.

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# Studies of multipole order in quadrupole Kondo lattice system PrV<sub>2</sub>Al<sub>20</sub> by low temperature thermal expansion and magnetostriciton measurement

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Recently, interesting phenomena, such as unconventional type of anomalous Hall effect and spin liquid like behaviour, have been observed in the Pr-based compounds [1, 2]. Among these compounds,  $PrTr_2Al_{20}$  (Tr = transition metal) have attracted much interest, because of the unique physical properties driven by the hybridization between conduction electrons and orbital (quadrupole) moments in the non-magnetic  $\Gamma_3$  ground doublet [3]. Such a hybridization is called quadrupole Kondo effect, and can induce non-Fermi liquid due to the overscreening.

 $PrV_2Al_{20}$  shows quadrupole ordering at  $T_Q = 0.6$  K and heavy fermion superconductor with  $T_C = 50$  mK at ambient pressure [3,4]. Above  $T_Q$ , non-Fermi liquid behaviour is observed, indicating the quadrupole Kondo effect. Moreover,  $T_Q$  is suppressed to zero by the magnetic field  $B_c \sim 11$  T parallel to [111], and non-Fermi liquid behaviour in electrical resistivity is observed at  $B_c$ , suggesting the possible field induced quantum critical point (QCP) [5]. Furthermore, the possibility which  $PrV_2Al_{20}$  has octupole order phase at low temperature and low field region is predicted [6].

To study the quantum criticality, Grüneisen coefficient  $\Gamma \sim \frac{\beta}{C} = -\frac{1}{VT} \frac{(\partial S/\partial p)_T}{(\partial S/\partial T)_p}$ , where  $\beta$  and C are

the volume thermal expansion coefficient and specific heat, is one of the most powerful probe because it diverges at any QCPs and shows universal critical scaling reflecting the type of QCP [7,8]. In this presentation, I will report the results of my thermal expansion and magnetostriction measurements of  $PrV_2Al_{20}$  at low temperature and discuss the possibility of quantum criticality and octupole order in this system.

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## Superconductivity in the laves phase SrIr<sub>2</sub>

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Recently, compounds based on 5d transition metals are attracting interests because novel physical properties are expected due to their strong spin-orbit coupling [1]. SrIr<sub>2</sub> is a superconductor containing 5d transition metal iridium in the laves phase and was discovered by Matthias *et al.* in 1957 [2]. Because there is no detailed report since then, the purpose of this research is to study the physical properties of SrIr<sub>2</sub> in detail.

We succeeded in synthesizing nearly single-phase SrIr<sub>2</sub>. It was found that SrIr<sub>2</sub> is a superconductor with  $T_c$  of 5.8 K (Fig. 1), which is slightly higher than the previously reported value of 5.7 K. We carried out the specific heat measurement and calculated superconducting parameters. It is suggested that SrIr<sub>2</sub> is a strongly coupled superconductor from the value of  $2\Delta(0)/k_BT_c \sim 4.26$  (>3.53 (BCS)). In this presentation, we report the results of X-ray structure analysis and also the superconducting parameters such as  $H_{c1}$  and  $H_{c2}$  determined from resistivity, specific heat and magnetization measurements.



**Fig.1.** Temperature dependence of the DC-magnetization in  $SrIr_2$  (ZFC and FC). The inset shows the expanded view around  $T_c$ .

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# Classification theory of odd-parity multipole order/superconductivity and electromagnetic responses

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Analysis of superconducting gap structure has been based on the group-theoretical classification by Sigrist and Ueda [1] for several decades. However, recent classification theory has revealed nontrivial gap structures beyond the Sigrist-Ueda theory. In this presentation, we show a comprehensive classification of " $j_z$ -dependent point nodes (gap opening)" [2]. Superconducting gap structure indeed depends on the angular momentum of Bloch states, in contrast to the prediction by the Sigrist-Ueda method. We suggest this unusual gap structure in heavy-fermion superconductors UPt<sub>3</sub>, UBe<sub>13</sub>, and PrOs<sub>4</sub>Sb<sub>12</sub>.

We also show a comprehensive group-theoretical classification of emergent multipole order in the condensed matter physics [3,4]. On the basis of the classification results in both real space and momentum space, we identify symmetry conditions and candidate materials for emergent electromagnetic responses. The responses include (1) piezoelectric effect, (2) magneto-piezoelectric effect, (3) Edelstein effect, (4) magneto-electric effect, (5) Antiferromagnetic spintronics, and (6) helical (FFLO) superconductivity.

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## Rock-salt-structured Chalcogenide Superconductor synthesized in high pressure

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Cubic chalcogenide, SnTe has been attracting interest after presenting the possibility of a topological crystalline insulator [1]. Doping In achieved the highest superconducting transition temperature ( $T_c$ ) around 4 K [2]. This is an enormous enhancement from 0.1 K of Sn<sub>1-x</sub>Te. In addition, surface sensitive spectroscopy revealed that Sn<sub>1-x</sub>In<sub>x</sub>Te shows topologically protected unique surface states [3]. This interest brought the material family of SnTe into the intensive studies, such as alloy compound with Pb, Pb<sub>1-x</sub>Sn<sub>x</sub>Te. The superconductivity of Sn<sub>1-x</sub>In<sub>x</sub>Te drew attention from another aspect, due to the limited valence state Sn, Pb and In show. This discontinuous valence state is proposed to be a possible mechanism of high  $T_c$  superconductivity [4].

In the course of aforementioned interests, we synthesized  $Sn_{1-x}In_xTe$  for the range of x>0.4. The conventional growth hit the solubility limit at x=0.45, however, high pressure synthesis enables the synthesis at higher x. We successfully synthesized for the compound of  $Sn_{1-x}In_xTe$  ( $x=0.5 \sim 1$ ) using the technique and confirmed the superconductivity in the range.  $T_c$  has a dome-like structure against x despite the monotonic decrease of lattice constant (Fig. 1(a, b)). The linear change of lattice constant and single phase synthesis indicate the successful doping of In in the system. Tc is enhanced in the range indicate that the superconductivity and is in good agreement with the calculated density of states behavior.



Fig. 1. (a) Lattice constant *a* increases along the increase of In doping (*x*). Data obtained in the work are shown as circles whilst the data in reference [5] is shown as squares. (b) Superconducting transition temperature ( $T_c$ ) against *x*. The maximum  $T_c$  sits in the vicinity of *x*=0.7.

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# de Hass-van Alphen effect of the itinerant weak ferromagnetic filled skutterudite LaFe<sub>4</sub>As<sub>12</sub>

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The filled-skutterudite compounds  $RTr_4X_{12}$  (R = rare earth; Tr = Fe, Ru and Os; X = P, As and Sb) which has the cubic LaFe<sub>4</sub>P<sub>12</sub>-type structure with space group  $Im\bar{3}$  have attracted much attention because of their novel physical properties. Among LaFe<sub>4</sub>X<sub>12</sub> compounds, LaFe<sub>4</sub>P<sub>12</sub> is a Pauli paramagnet with the electronic specific heat coefficient  $\gamma$  = 57 mJ/K<sup>2</sup>mol,<sup>1</sup> while LaFe<sub>4</sub>As<sub>12</sub> and LaFe<sub>4</sub>Sb<sub>12</sub> show Curie-Weiss behaviors and have larger values of  $\gamma$  = 135 mJ/K<sup>2</sup>mol and 200 mJ/K<sup>2</sup>mol, respectively.<sup>2,3</sup> In order to understand these unusual electronic states, we tried to grow high quality of single crystals and measured the de Hass-van Alphen (dHvA) effect of LaFe<sub>4</sub>As<sub>12</sub> and revealed the Fermi surface properties.

High quality samples of LaFe<sub>4</sub>As<sub>12</sub> with residual resistivity ratio of 280 have been synthesized under high pressure of 4GPa at 1000°C. The dHvA measurements have been performed by two different methods of a modulation coil method and canti-lever method using a top loading dilution refrigerator down to 60 mK with a 17 T superconducting magnet. The angular dependence of the dHvA frequency in LaFe<sub>4</sub>As<sub>12</sub> was measured every 4.5 degrees for whole principal field direction. The angular dependence of dHvA indicates that the Fermi surface of LaFe<sub>4</sub>As<sub>12</sub> consists of a closed sheet and a multiply connected one. The topology and the size of those Fermi surfaces are basically in good agreement with a band calculation. It is also revealed that the cyclotron effective mass  $m^*$  is roughly two times larger than the calculated band mass.

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Fig. 1. The angular dependence of dHvA frequency of  $LaFe_4As_{12}$ .

# Single Crystal Growth and Structural Analysis of Ternary Compound *R*-Ir-Sn (*R*: rare earth) with a Large Unit Cell

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The unconventional electronic states appearing in chiral and noncentrosymmetric structures have attracted much attention in the field of condensed-matter physics. According to the grouptheoretical classification of electronic states, orbital Weyl points appear intrinsically at particular points on the Brillouin-zone boundary for chiral space groups, which give rise to the bulk chiral fermion and the Weyl semimetal state. Therefore, the investigation of chiral-structure systems has become a important research subject.

Recently we focus on the investigation of the 3-4-13 and related materials, especially for *R*-*Tr*-Sn system (R: rare earth, Tr: transition metal). The details of R-T-Sn ternary phases have been reported by G. P. Espinosa and A. S. Cooper, and their coauthors in Bell Labs since 1980 's. Some of compounds have a relatively large unit cell structure and a complex and various phases [1-3]. In these systems,  $R_3T_4Sn_{13}$  series are well known as compounds which are relatively easy to exhibit superconductivity. Although this series has a several crystal structures, one of the most major structures is the Yb<sub>3</sub>Rh<sub>4</sub>S<sub>13</sub>-type cubic structure as shown in Fig. 1 [4]. It was reported that a some of  $R_3T_4Sn_{13}$  have the structural instability and show a phase transition from a primitive cubic to a body centred cubic structure with chirality [5]. Therefore, we have tried to do a systematic single crystal growth on rare earth elements and investigated their physical properties. As a result, it is gradually clarified the details of the crystal structures of those crystals and a crystallographical relation to a different phase of these systems. For example, although the relative atomic position is basically same as Yb<sub>3</sub>Rh<sub>4</sub>S<sub>13</sub>-type in Fig. 1, it has a tetragonal structure and unit cell volume takes a rational number multiple. Recently we also succeeded in growing single crystals of La<sub>3</sub>Ir<sub>4</sub>Sn<sub>13</sub> and Lu<sub>5</sub>Ir<sub>6</sub>Sn<sub>18</sub> as shown in Fig. 2 by Sn self-flux method. We tried to identify the elemental composition of those single crystals by using EDX and the single crystal XRD. The detailed structural parameters were determined as a cubic and a tetragonal structure, respectively. For La<sub>3</sub>Ir<sub>4</sub>Sn<sub>13</sub>, the space group of the room temperature phase can be identified as a chiral cubic  $I4_13_2$ . We will report details of physical properties of these compounds.



Fig.1. Yb<sub>3</sub>Ir<sub>4</sub>Sn<sub>13</sub>-type cubic structure.[4]

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# Slave-boson mean-field study of multi-orbital periodic Anderson model

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The archetypal heavy fermion superconductor, UBe<sub>13</sub> has attracted much interest in many years due to the non-Fermi-liquid (NFL) behavior in the normal state as well as the unconventional properties in the superconducting phase [1]. Cox proposed the quadrupolar Kondo effect as a possible origin of the NFL behavior in UBe<sub>13</sub> based on the assumption that the non-Kramers crystalline-electric-field (CEF) doublet ground state with  $\Gamma_3$ -symmetry is realized on each U-site [2]. Recent experiments for Th<sub>1-x</sub>U<sub>x</sub>Be<sub>13</sub>, however, imply that the  $\Gamma_1$ -singlet state is another promising candidate of CEF ground state and the NFL behavior is due to the competition between CEF- and Kondo-singlets [3,4].

Although the impurity Kondo problem with  $f^2$ -singlet CEF ground state has been theoretically explored by many authors [5,6], there are few studies on its lattice version [7,8]. In the present study, we investigate the competition between CEF- and Kondo-singlets in the multi-orbital periodic Anderson model on the simple cubic lattice, where we take into account *f*-orbitals with total angular momentum j=5/2 and conduction *d*-orbitals with  $t_{2g}$ -symmetry. The effects of Coulomb interaction are treated within the rotationally invariant slave-boson mean-field approximation (SBMFA) [9].

Figures 1(a) and (b) show the *c-f* hybridization ( $V_{cf}$ )-dependence of the quasi-particle occupation and effective *f*-level. For  $V_{cf} < 0.56$ , the CEF-singlet state is realized, while for  $V_{cf} > 0.57$ , the Kondosinglet state is realized. Within the SBMFA, the change between CEF- and Kondo-singlet states is 1st-order transition, consistently with the previous study based on a more simplified model [8]. As shown in Figs. 1(c) and (d), across the transition point, Fermi surfaces (FSs) drastically change since in the Kondo-singlet state, *f*-electrons participate to form the FSs, while in the CEF-singlet state, they do not. It should be noted that the transition is not accompanied by any symmetry breaking and both

singlet states are paramagnetic. The *f*-electron part of multipolar susceptibility  $\chi(q)$  also shows distinct behavior in each singletphase. In the Kondo-singlet phase,  $\chi(q)$  has moderate *q*dependence originating from the particle-hole excitation near the Fermi level. On the other hand, in the CEF-singlet state,  $\chi(q)$  is almost flat in *q*-space since there are only Van Vlecktype contributions to  $\chi(q)$ .



Fig. 1.  $V_{cf}$ -dependence of (a) quasi-particle occupation and (b) effective *f*-level measured from the Fermi level. (c) and (d) Fermi surfaces for CEF- and Kondo-singlet states, respectively.

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# Single crystal growth and low-temperature physical property measurements of quadrupolar Kondo lattice $PrTr_2Al_{20}$ (Tr = Mo, W)

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 $\Pr Tr_2Al_{20}$  (Tr = transition metal) compounds have attracted much attention as new candidates for investigating the quadrupolar (two-channel) Kondo effect because  $\Pr Tr_2Al_{20}$  (Tr = Ti, V) show Kondo effect and superconductivity attributed to quadrupolar degrees of freedom[1]. Recently, we have investigated  $\Pr Tr_2Al_{20}$  (Tr = Nb, Ta) single crystals and pointed out the possibility that quadrupole Kondo lattice are also realized in these compounds[2]. In order to investigate the electronic state of  $\Pr Tr_2Al_{20}$  systematically, we have tried to grow higher-quality single crystals of  $\Pr Tr_2Al_{20}$  (Tr = Mo, W) and performed physical property measurements.

The result of single-crystal X-ray structural analysis for  $PrMo_2Al_{20}$  (Fig. 1) shown in Table 1 indicates the existence of deficiency in the Mo site. On the other hand, as a result of the composition analysis using SEM-EDX, the element ratio of averaged Pr, Mo, and Al within the beam size range was approximately 1:2:20. The consistency of these two results indicates a possibility of element substitution of Mo for Al. Based on this interpretation, we optimised conditions of growing crystals.

We have also performed specific heat measurements to clarify the ground state of  $PrTr_2Al_{20}$  (Tr = Mo, W). C/T of  $PrTr_2Al_{20}$  (Tr = Mo, W) does not show any anomaly indicating a phase transition down to 0.4 K and has a large Sommerfeld coefficient of 0.7-1.0 J/mol K<sup>2</sup> at low temperatures. In contrast to  $PrTr_2Al_{20}$  (Tr = Nb, Ta), there is broad peak around 2 K that shifts to higher temperature with increasing magnetic fields.  $S_{4f}$  saturates around Rln5 at 50 K without a clear plateau at Rln2. In this poster, we will show recent results for  $PrTr_2Al_{20}$  (Tr = W, Mo) and discuss the possibility of the realization of quadrupolar Kondo lattice in these compounds.



Atom	site	x	у	Z	$B_{\rm eq}({\rm \AA}^2)$	occ
Pr	8 <i>a</i>	1/8	1/8	1/8	0.727(12)	1
Mo	16 <i>d</i>	1/2	1/2	1/2	0.489(14)	0.888
Al(1)	96g	0.059	0.059	0.32497	0.864(14)	1
Al(2)	48 <i>f</i>	0.48688	1/8	1/8	0.868(18)	1
Al(3)	16 <i>c</i>	0	0	0	1.54(3)	1

Fig. 1. Single Crystals of PrMo<sub>2</sub>Al<sub>20</sub>

Table 1. Atomic coordinates and thermal parameters of PrMo2Al20.  $R_1 = 1.19$  %,  $wR_2 = 2.63$  %

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# The Effect of Doping on the Transport Properties of the Non-collinear Antiferromagnet Mn<sub>3</sub>Sn

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Below its  $T_N \sim 420$  K, the hexagonal compound Mn<sub>3</sub>Sn orders in triangular spin structure which exhibits a vanishing in-plane magnetization of ~  $3m\mu_B/Mn$  and small coercivity of ~ 200 Oe . Interestingly, the electronic band structure associated with this magnetic ordering contains multiple Weyl nodes, the closest of which is around 60 meV above the Fermi energy, resulting in large anomalous Hall conductivity [1] and Nernst effect [2] driven by the Berry curvature of the Weyl nodes. Recently, ARPES measurement and the detection of the chiral anomaly in this compound lend some evidence to the existence of Weyl nodes around the Fermi energy. Due to the distribution of the Berry curvature in the momentum space, the anomalous transport properties of Mn<sub>3</sub>Sn are predicted to be highly sensitive to chemical potential change induced by carrier doping. On the other hand, it also has been known that despite being independent of the scattering time, Berry curvature induced transport properties can also be suppressed by sufficiently strong disorder [4]. Here we will talk about the transport and magnetic properties of Mn<sub>3</sub>Sn single crystals doped with various elements and discuss them in relation to the theoretical calculation.

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## NMR Study of GaTa<sub>4</sub>Se<sub>8</sub>

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Cubic GaTa<sub>4</sub>Se<sub>8</sub> is a magnetic cluster system, in which a Ta<sub>4</sub> unit (tetramer) possesses the magnetic moment. It shows insulating behavior and has been classified as Mott insulator. This compound shows various interesting properties; pressure-induced Mott transition, pressure-induced superconductivity, a phase transition at 53 K, *etc.*.[1-3] However, contributions of spin-orbit coupling for Ta-5d electrons to these phenomena are not clarified yet.

We synthesized small single crystals using the same manner as the previous report.[4] Figure 1 shows the temperature dependence of magnetic susceptibility of GaTa<sub>4</sub>Se<sub>8</sub>. Above  $T_S = 53$  K, it shows Curie-Weiss like behavior, and a sudden drop appears below  $T_S$ . An origin of a similar phase transition in isostructural GaNb<sub>4</sub>S<sub>8</sub> has been suggested to be a formation of Nb<sub>8</sub> octamer from NMR measurements.[5] In this study, we performed NMR measurements on GaTa<sub>4</sub>Se<sub>8</sub> to clarify the electronic state. Figure 2 shows <sup>77</sup>Se-NMR spectrum at ~2 T. In the high-temperature phase above 53 K, the cubic structure has two inequivalent Se sites; therefore, two signals denoted as Se1 and Se2 are observed. The two signals suddenly shift below 53 K because of the reduction of susceptibility. The spectral shape at 53 K indicates that the transition is accompanied by a phase separation, indicative of the first-order phase transition. Below 53 K, the spectral width is relatively broader irrespective of the small susceptibility. To distinguish whether the broadening originates in the distribution of the orbital part of Knight shift or the distribution of the internal field, we are checking the field dependence of the spectrum. We also report the nuclear spin-lattice relaxation rate and some results of Ga-NMR.



g. 1 Temperature dependence of GaTa<sub>4</sub>Se<sub>8</sub>

Fig. 2<sup>77</sup>Se-NMR spectrum of GaTa<sub>4</sub>Se<sub>8</sub>

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## NMR-study on the artificial superlattice CeCoIn<sub>5</sub>/CeRhIn<sub>5</sub>

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Magnetic fluctuations in strongly correlated electron systems have been intensively studied from experimental and theoretical aspects, since most of unconventional superconductors have been discovered in the verge of the magnetic phase and antiferromagnetic possess strong (AFM) heavy-fermion fluctuations. The (HF) superconductor CeCoIn5 is one of the such unconventional SCs. The superconducting (SC) transition temperature  $T_c$  of CeCoIn<sub>5</sub> is 2.3 K[1], which is the highest  $T_c$  among Ce-based HF superconductors. In addition, it has been considered that the superconductivity is mediated by AFM spin fluctuations with quantum critical character.

Recently, progress in the epitaxial-growth technique enabled us to synthesize an artificial HF superlattices (SLs) of alternating block layers (BLs) with a few atomic layer thickness[2]. It is reported that a SL CeCoIn<sub>5</sub> (HF superconductor)/ CeRhIn<sub>5</sub> (spin-density-wave metal) shows enhancement of the Pauli-limited upper critical field, indicative of



increase of superconducting gap  $\Delta$  under pressure[3]. The SL provides new platform to study relationship between HF superconductivity and magnetic interactions.

We performed <sup>59</sup>Co-nuclear magnetic resonance (NMR) measurement focused on the CeCoIn<sub>5</sub> BL in CeCoIn<sub>5</sub>(5)/ CeRhIn<sub>5</sub>(5) SL[the number in () is the number of unit-cell-layers of each BL.] to investigate how spin fluctuations are changed by magnetic interaction between both BLs. We found a larger  $1/T_1T$  component than that of the CeCoIn<sub>5</sub> films shows up with approaching to the magnetic order temperature  $T_N$  in the CeRhIn<sub>5</sub> BL as shown in the Fig., although the same  $1/T_1T$  component arising from the inner layers of the CeCoIn<sub>5</sub> BL remained. It is suggested that AFM spin fluctuations at the interfaces are enhanced by magnetic interaction with the adjacent CeRhIn<sub>5</sub> BL.

At present, we are studying  $CeCoIn_5(7)/CeRhIn_5(7)$  SL to investigate BL-thickness dependence of spin fluctuations.

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# NMR study on dynamical property of tetrahedrite Cu<sub>12</sub>Sb<sub>4</sub>S<sub>13</sub>

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Tetrahedrites  $Cu_{10}Tr_2Sb_4S_{13}$  (Tr = transition metal), which is a potential candidate for thermoelectric transducer, has attracted attention because this system exhibits anomalous physical properties at low temperatures[1].

Figure 1 shows the crystal structure of tetrahedrite. The crystal structure belongs to a bodycentered-cubic (bcc) system with space group *I*-43*m* (No. 217, Z = 2) at room temperature. The complex crystal structure contains 58 atoms in the unit cell. There exist two Cu sites named as Cu(1) and Cu(2) and also two S sites named as S(1) and S(2). Cu(1) site is tetrahedrally surrounded by four S(1) atoms and Cu(2) site is trigonally surrounded by two S(1) atoms and one S(2) atom. It is reported that Cu(2) atom has large atomic displacement parameter along the axis perpendicular to S(1)<sub>2</sub>S(2) trigonal plane[2,3]. This fact means that Cu(2) has a vibration mode with large amplitude oscillation such as "rattling", although tetrahedrite does not have cage structure like as clathrates which have guest-host structure.

We have studied dynamical property of vibration mode of Cu atom by NMR relaxation rates  $1/T_1$ , where  $T_1$  reflects information of magnetic/electric fluctuations. We measured  $T_1$  of Cu(1) nuclear for various magnetic fields H = 7 T, 8.87 T, 12 T and found that  $1/T_1$  obeys following formula,  $1/T_1 = \Delta^2 \omega / (\omega^2 + \omega_N^2)$ , where  $\Delta$  is transition matrix element,  $\omega$  is frequency of fluctuation,  $\omega_N$  is NMR frequency. Furthermore, the temperature dependence of  $1/T_1$  can be explained when  $\omega$  has the Arrhenius-type temperature dependence  $\omega = A \exp(-E/k_B T)$  with activation energy *E*. The activation energy is estimated as E = 0.16 eV. Using this gap parameter, we also explained the temperature dependence of  $T_1$  of Cu(2) site. We will discuss about these results in this conference.



Fig. 1. Crystal structure and  $S(1)_2S(2)$  plane of tetrahedrite  $Cu_{12}Sb_4S_{13}$ . Ellipsoid reflects atomic displacement parameter[2].

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# NMR Study of CeCu<sub>2</sub>Si<sub>2</sub> under High Field and Pressure

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CeCu<sub>2</sub>Si<sub>2</sub> shows anomalous increase in  $T_{\rm C}$  around the critical pressure  $P_{\rm C} = 4.5$  GPa. It is proposed that superconducting mechanism is mediated by the valence fluctuation of Ce ions in high pressure phase. Our previous study revealed that the <sup>63</sup>Cu-NQR frequency (<sup>63</sup> $v_{\rm Q}$ ) suddenly deviated from the linear pressure dependence of <sup>63</sup> $v_{\rm Q}$  above 4 GPa, suggesting that Ce valence rapidly increases around  $P_{\rm C}$ . [1, 2] Since valence transitions are not observed in powdered XRD measurement under pressure, valence crossover occurs in the narrow pressure range around  $P_{\rm C}$ . Recently, theory of valence mediated superconductivity proposed that the (static) valence transition appears at finite temperature with applying high magnetic field. [3] Therefore we measured Cu-NMR of single crystal CeCu<sub>2</sub>Si<sub>2</sub> in order to elucidate the field and pressure dependence of Ce valence and spin state.

Three sharp <sup>63</sup>Cu-NMR lines were observed, suggesting that our single crystal is of good quality. <sup>63</sup>Cu-NQR frequency (<sup>63</sup> $v_Q$ ) was determined from two satellite lines. In the ambient pressure, <sup>63</sup> $v_Q$  is almost temperature independent below 30 K and whole *T*-dependence is magnetic field independent up to 17 T. Above  $P_C$ , <sup>63</sup> $v_Q$ 's for P = 5.4 GPa and 6.2 GPa, decrease clearly below  $T^* = 20$  K and 40 K at 18 T, respectively. Knight shift (*K*) and nuclear relaxation rate (1/ $T_1$ ) for <sup>63</sup>Cu nuclei also decrease clearly below  $T^* = 20$  K and 40 K at 18 T, respectively. Knight shift (*K*) and nuclear relaxation rate (1/ $T_1$ ) for <sup>63</sup>Cu nuclei also decrease clearly below  $T^* = 20$  K and 40 K for 5.4 GPa and 6.2 GPa, respectively. Density of state at Fermi level decreases below  $T^*$  simultaneously with decrease of electric field gradient. These new findings suggest that some (valence and/or orbital) crossover occurs around  $T^*$  under high pressure.  $T^*$  seems to approach down to zero at the critical pressure  $P_C = 4.5$  GPa. Furthermore, we found that the crossover temperature was field independent between 5 and 22 T at 6.2 GPa. More detailed high pressure experiments under the magnetic field are needed to elucidate the nature of new crossover behavior.

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# Magnetic Properties and Magnetic Phase Diagram of Single Crystalline YbNiSn

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A ferromagnetic superconductor URhGe with TiNiSi-type orthorhombic crystal structure shows quite unique phenomena which are the spin reorientation and the reentrant superconductivity for  $H \parallel b$ -axis [1, 2]. In order to understand the collapse of the Curie temperature under magnetic field, we have studied a heavy fermion ferromagnet YbNiSn with the identical crystal structure. YbNiSn is a heavy fermion compound with the Curie temperature  $T_c=5.5$  K and Sommerfeld coefficient  $\gamma=300$  mJ/(K<sup>2</sup>mol) [3-5]. We have succeeded in growing high-quality single crystals using the Bridgman method with W-crucibles. In the present work, we clarify the highly-anisotropic magnetic properties and its magnetic phase diagram by measuring the electrical resistivity, magnetization, and thermoelectric power.

The residual resistivity  $\rho_0$  is 1.37  $\mu\Omega$ ·cm and the residual resistivity ratio RRR (=  $\rho_{RT}/\rho_0$ ,  $\rho_{RT}$ : resistivity at room temperature) is 55, revealing a high-quality sample. The electrical resistivity  $\rho(T)$  shows a double-peak structure around 60 K and 10 K with Kondo-lattice behavior as same as a previous study [3], and  $\rho(T)$  exhibits a drop due to the ferromagnetic transition at  $T_C = 5.68$  K. We also measured a magnetic field dependence of the electrical resistivity of YbNiSn for  $H \parallel a$ -axis. The electrical resistivity under magnetic fields show plateau in a field range from 10 to 20 kOe below 5 K due to a spin-flop. With increasing temperature, the field range of the resistive plateau becomes narrower and finally disappears above  $T_C$ . A field dependence of the magnetization M for  $H \parallel a$ -axis shows the canting process and three anomalies at the fields  $H_{c1}$ ,  $H_{c2}$ , and  $H_{c3}$ . Such phenomena come from the spin-flop behaviors for an Ising ferromagnet under magnetic fields in agreement with the previous studies [4, 5]. The values of  $H_{c1}$ ,  $H_{c2}$ , and  $H_{c3}$  decrease with increasing temperature and merge at around the Curie temperature. Finally, we summarize the magnetic phase diagram of YbNiSn for  $H \parallel a$ -axis and discuss the nature of anomalies observed from our transport and magnetization measurements.

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## X-ray Crystal Structure Analysis of Single-Crystalline UNi<sub>4</sub>B

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

A small piece (about 20 micron in diameter) of single crystalline UNi<sub>4</sub>B grown by Czochralski method was used as a specimen. The diffraction of synchrotron X-ray was measured by using a diffractometer with an imaging-plate type detector at a beamline PF-AR NE1A in Photon Factory. The high energy X-ray of 30 keV was utilized to reduce the large absorption effect of the heavy U atoms in the sample. The obtained diffraction patterns strongly suggest the orthorhombic unit cell, whose lattice constants are: a = 6.922(4) Å, b = 14.773 (2) Å, c = 17.04 (1) Å, which is the same as the one of the previously reported structure. The direct method using SIR2011 gives a structure in which U atoms form distorted trianglar lattices, without local inversion symmetry at each U site. This is very important information because according to the above theory, realization of toroidal order requires that the sites of magnetic ions have no local inversion symmetry. In the presentation, we will discuss possible structure of the toroidal order realizing in UNi<sub>4</sub>B from the viewpoint of detailed local crystallographic symmetry at magnetic U sites.

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# Magnetic Phase Transitions in Single Crystalline Non-centrosymmetric URhSn under Pressure

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We report the effect of hydrostatic pressure on a well characterized single crystal of URhSn, which crystallizes in a non-centrosymmetric ZrNiAl- type structure with the U-atoms forming a quasi-kagome lattice. URhSn is known to exhibit magnetic transitions at  $T_1 = 54$  K and  $T_2 = 17$  K with a ferromagnetic ground state [1,2,3]. We have measured electrical transport in the c-plane of single crystalline URhSn in a Bridgman anvil cell attached with a lead manometer to explore the evolution of these phase transitions up to a pressure of 5.9 GPa and temperatures down to 3 K. We find that the  $T_1$  is monotonically suppressed to 37 K, while the  $T_2$  first decreases at 1 GPa, shows a maximum and attains a value of 18 K with a declining trend at 5.9 GPa (Fig. 1). From the temperature-pressure phase diagram, a critical pressure of ~ 8 GPa is anticipated. A least square fitting of the equation  $\rho = \rho_0 + AT^2$  to the resistivity vs. temperature data in temperature range 3-5 K reveals a diverging trend in the *A* coefficient as well as in the residual resistivity ( $\rho_0$ ). This indicates an enhancement in the quasiparticle mass and presence of a possible quantum critical point at a higher pressure in URhSn.



Fig. 1. Variation of the magnetic transition temperatures, the A coefficient, and the residual resistivity of URhSn under pressure.

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## Thermoelectric properties of a Weyl magnet thin film

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Over the past few years, the Weyl semimetal (WSM) has attracted considerable interest in the field of condensed matter physics. In WSMs, due to either inversion symmetry breaking or timereversal symmetry (TRS) breaking, two nondegenerate energy bands can linearly touch at pairs of isolated points in the momentum space, called as Weyl points. The touching points in the momentum space act as the (anti) monopole of fictitious field, that is Berry curvature, giving rise to the topological properties. Especially, WSMs realized by TRS breaking due to the magnetic properties are called as Weyl magnets. WSMs show many exotic bulk properties such as the anomalous Hall effect (AHE), the anomalous Nernst effect (ANE), the magneto-optical Kerr effect (MOKE), and chiral anomalies, which can be realized by Berry curvature. WSMs realized by inversion symmetry breaking have been reported experimentally [1], but Weyl magnets have been reported only theoretically. However, it has been experimentally reported that Mn<sub>3</sub>Sn is a Weyl magnet for the first time. Mn<sub>3</sub>Sn is a hexagonal antiferromangnet with a space group of P6<sub>3</sub>/mmc that exhibits noncollinear ordering of Mn magnetic moments at the Néel temperature of  $T_N \sim 420$  K [2]. In the bulk system, Mn<sub>3</sub>Sn shows the large AHE, ANE and MOKE at room temperature, which is comparable with those of ferromagnets [3-5]. Besides these results, the analysis of the band structure calculated by density functional theory and measured by angle-resolved photoemission spectroscopy, and the observation of the chiral anomaly by magnetotransport measurements provide evidence for the existence of magnetic Weyl fermion in Mn<sub>3</sub>Sn [6].

Moreover, ANE has attracted a great deal of attention as one of some thermoelectric effects of the direct and environment-friendly energy conversion from heat to electricity. In ANE, the Nernst voltage will be induced normal to spontaneous magnetization and thermal current which are perpendicular to each other. The fabrication process to make a lateral thermopile structure of ANE-based thermoelectric power generation is easier than of Seebeck effect-based thermoelectric power generation. Antiferromagnets produce negligible stray field and are robust against perturbation due to magnetic field, so the devices using antiferromagnets are suitable for high-density integration. Thus, when antiferromagnets show ANE, ANE-based thermoelectric generation using them can be applicable. In order to achieve this, we need to produce antiferromagnetic thin films suitable for flexible fabrication. Thus, we synthesized Mn<sub>3</sub>Sn thin films.

We tried to synthesize Mn<sub>3</sub>Sn thin films, and succeeded in making the polycrystalline thin film which has the only phase of Mn<sub>3</sub>Sn. We found that Mn<sub>3</sub>Sn thin films exhibit a small net magnetization of several m $\mu_B$ . This result is the same as that of Mn<sub>3</sub>Sn bulk system. Moreover, we measured the thermoelectric properties as a function of magnetic field, when thermal current was applied in the orientation of out-of-plane and magnetic field in the orientation of in-plane. We observed the thermal electromotive force originated from ANE, where the sign change as a function of field comes from the flipping of the spin texture. We will report the measurement method and results of the thermoelectric properties of Mn<sub>3</sub>Sn thin films.

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# NMR Spectrum Analysis and High-Field Magnetization in PrT<sub>2</sub>Al<sub>20</sub> (T=Nb, Ta)

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Intermetallic compounds  $RT_2X_{20}$  (*R*: rare earth, *T*: transition metal, *X*: Al, Zn, and Cd) attract much attention because of their various intriguing phenomena such as ultra-heavy fermion state [1], quadrupolar Kondo effect [2,3], field insensitive heavy fermion state [4,5] at low temperatures.

Among them,  $\Pr T_2 X_{20}$  system shows quadrupole order, unconventional superconductivity, and non-Fermi liquid behaviors. The origin of these novel behaviors is believed to be strong c-fhybridizations between conduction electrons and the  $\Gamma_3$  CEF ground state of 4*f* electrons.  $\Pr T_2 X_{20}$  is believed to be an ideal playground to study the quadrupolar Kondo effect because of the coexistence of strong *c*-*f* hybridizations and the higher order multipoles.

 $PrT_2Al_{20}$  (*T*: Nb, Ta) show the NFL behaviors in the electric resistivity  $\rho$  and magnetization below 5 K [6,7]. PrNb<sub>2</sub>Al<sub>20</sub> shows no long range ordering down to 0.075 K [8] and PrTa<sub>2</sub>Al<sub>20</sub> shows an antiferroquadrupole ordering at 0.65 K [7]. The *T*-dependence of  $\rho$  between 1 and 5 K is well reproduced by the quadrupolar Kondo lattice model [9].

In the present study, we performed <sup>27</sup>Al- and <sup>93</sup>Nb-NMR measurements using powder samples and a single crystal of  $PrT_2Al_{20}$  (*T*: Nb, Ta). Figure 1 shows the temperature dependence of <sup>93</sup>Nb isotropic Knight shift  $K_{iso}$  and linewidth  $\sigma$  in  $PrNb_2Al_{20}$  obtained at 8.57 T with  $H \parallel \langle 111 \rangle$ .  $K_{iso}$  and  $\sigma$ are obtained by analyzing the entire NMR spectrum. *K* and  $\chi$  have good linearity and we could obtain the field dependence of local magnetization  $M_{loc}$  for 3–19 T at 2 K. Meanwhile,  $\sigma$  suddenly increases below 40 K as the temperature is lowered. Such a broadening might originate from the distribution of the field-induced magnetic dipoles because the <sup>93</sup>Nb-NQR linewidth shows no temperature dependence [8].

We will report the analysis of NMR line profile and the temperature and field dependence of  $K_{iso}$ ,  $\sigma$ , and  $M_{loc}$  and will discuss the static 4*f* electronic

state in  $PrT_2Al_{20}$  from microscopic viewpoints.

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Fig. 1. Temperature dependences of  $K_{iso}$  and  $\sigma$  in PrNb<sub>2</sub>Al<sub>20</sub> single crystal with  $H \parallel \langle 111 \rangle$ .