

氏名	ヤマダ アキラ 山田 瑛
所属	理工学研究科 物理学専攻
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学位論文題名	Unusual Magnetic-Field-Insensitive Heavy Fermion States in Sm T_2 Al $_{20}$ (T : Transition Metals) Sm T_2 Al $_{20}$ (T :遷移金属)における異常に磁場鈍感な重い電子状態 (英文)
論文審査委員	主査 教授 青木 勇二 委員 准教授 松田 達磨 委員 准教授 服部 一匡

【論文の内容の要旨】

Strongly correlated f electron systems have attracted much attention due to a variety of physical phenomena, such as heavy fermion (HF), non-Fermi liquid (NFL) and unconventional superconductivity. These behaviors originate from the degrees of freedom possessed by f electrons, such as magnetic dipole and higher multipole moments associated with crystalline electric field (CEF) states, and possible multiple valence states. So far, studies have been made intensively in Ce-based compounds and, for the other rare-earth elements, they remains to be elucidated. Unique behaviors that have been found recent years are unconventional quantum critical behaviors probably driven by valence instabilities in YbRh $_2$ Si $_2$ and β -YbAlB $_4$, quadrupolar Kondo effect and associated NFL behaviors in Pr T_2X_{20} (T : transition metals, X : Al, Zn, and Cd). For Sm, only a limited number of strongly correlated behaviors have been reported. Among them, "unusually magnetic-field-insensitive HF states" observed in SmOs $_4$ Sb $_{12}$ and Sm T_2 Al $_{20}$ (T : Ti, V, and Cr) are one of the important topics remaining to be elucidated. Unconventional non-magnetic mechanisms are expected to be involved in these phenomena. In order to resolve this issue, we focus on the Sm T_2 Al $_{20}$ systems. We have grown single crystals of Sm T_2 Al $_{20}$ and other relevant materials, and have performed

single-crystal X-ray analysis and electrical transport, magnetization, specific heat, and X-ray absorption spectroscopic (XAS) measurements. Main findings are summarized below.

1. Heavy fermion states unusually insensitive to magnetic field in $\text{SmTa}_2\text{Al}_{20}$

We have succeeded in growing single crystals of $\text{SmTa}_2\text{Al}_{20}$ and have measured the physical properties. It has been found that a magnetic-field-insensitive phase transition appears at 2.0 K and the electronic specific heat coefficient is $\gamma \cong 3 \text{ J/mol K}^2$, largest among Sm-based intermetallic compounds. Specific heat data suggest that the CEF ground state of the $\text{Sm}^{3+} J=5/2$ multiplet is a Γ_8 quartet. The size of the ordered Sm dipole moment is estimated to be $0.22 \mu_B/\text{Sm}$ from nuclear specific heat in zero magnetic field, which is suppressed compared with those expected for a Γ_8 ground state. The Sm L_3 -edge XAS measurements have clarified that Sm ions are in an intermediate valence state with the average valence of about +2.85 with no significant temperature dependence. This is in line with the observed temperature dependence of magnetic susceptibility, which is evidently different from those of both free Sm^{2+} and Sm^{3+} ions.

The resistivity shows a clear $-\log T$ dependence, suggesting the occurrence of Kondo effect. The transverse magnetoresistance (MR) data show a strong deviation from the Kohler's rule in the low-field condition, indicating that the conduction electron scattering by $4f$ electrons has a strong wave-vector dependence. Analyses clarified that the MR data obey "the modified Kohler's rule" that has been confirmed to be satisfied in some of the quasi-two-dimensional strongly-correlated electron systems. This finding, providing the first example in cubic systems, reveals that the modified Kohler's rule can be applied to three-dimensional systems.

2. Single-site Kondo effect in $\text{Sm}_x\text{La}_{1-x}\text{Ta}_2\text{Al}_{20}$

The La-substitution effect on the transport properties has been examined using single crystals of $\text{Sm}_x\text{La}_{1-x}\text{Ta}_2\text{Al}_{20}$ ($x = 0.01, 0.05, 0.25, \text{ and } 1$). The magnetic-field-insensitive $-\log T$ dependence of resistivity has been observed even in the most dilute sample with $x = 0.01$, clearly evidencing that the unconventional $-\log T$ dependence is caused by a local single-ion Kondo effect. The transverse MR of $\text{Sm}_x\text{La}_{1-x}\text{Ta}_2\text{Al}_{20}$ is always positive with H^2 dependence. The deviation from the

Kohler's rule gradually becomes large as the Sm concentration x increases, indicating that the strong wave-vector dependent conduction electron scattering is caused by Sm ions. The XAS measurements down to $x = 0.01$ clarify that the average Sm-ion valence is always +2.85 and independent on temperature and x , suggesting that this is a Sm single-ion character.

3. Superconducting properties in $\text{La}Tr_2\text{Al}_{20}$ (Tr : Ti, V, Nb, and Ta)

To investigate the characteristics of conduction electrons that host the above-mentioned strongly correlated electron states, the physical properties of reference materials $\text{La}Tr_2\text{Al}_{20}$ (Tr : Ti, V, Nb, and Ta) have been measured. It has been found that all these are superconductors with the transition temperature $T_c = 0.46, 0.18, 1.05, \text{ and } 1.02$ K, respectively. Analyses using the modified McMillan formula suggest that these are weak-coupling type-II superconductors. Single crystal X-ray diffraction analysis indicates a large atomic displacement parameter at Al (16*c*) site, suggesting "rattling" anharmonic large-amplitude oscillations of the Al ions. No such feature is found for the atomic displacement parameter at La site on the cage center, in contrast to the rattling cage-center ions in $\text{Ga}_{0.2}\text{V}_2\text{Al}_{20}$, $\text{Al}_{0.3}\text{V}_2\text{Al}_{20}$, $\text{ScV}_2\text{Al}_{20}$, and $\text{LuV}_2\text{Al}_{20}$. The large T_c distribution in the four $\text{La}Tr_2\text{Al}_{20}$ compounds indicates that anharmonic large-amplitude vibration modes of Al ions at 16*c* site and/or d electrons of transition metals may play an essential role for the Cooper pairing.

4. Low-Curie temperature ferromagnetic phase in $\text{SmPt}_2\text{Cd}_{20}$

In $\text{Sm}Tr_2X_{20}$ compounds (Tr : transition metals, X : Zn and Cd), the $4f$ electronic states are relatively localized. This feature can be seen in the clear Curie-Weiss behavior in temperature dependence of magnetic susceptibility and the absence of the Kondo scattering ($-\log T$ dependence) in resistivity. We have succeeded in growing single crystals of $\text{SmPt}_2\text{Cd}_{20}$, a new member of $\text{Sm}Tr_2X_{20}$ family. It has been found that $\text{SmPt}_2\text{Cd}_{20}$ exhibits a ferromagnetic (FM) transition at $T_{\text{FM}} = 0.64$ K, which is the lowest among the cubic 1-2-20 compounds. The increase of specific heat divided by temperature with decreasing temperature even below T_{FM} and a power-law behavior with $T^{0.74}$ dependence in resistivity below 2 K

imply substantial magnetic quantum fluctuations. An analysis of the magnetic entropy suggests the CEF splitting of the $J=5/2$ multiplet of Sm^{3+} state with a Γ_7 doublet ground state and a Γ_8 excited state with the excitation energy of ~ 30 K. The bulk properties at low temperatures suggest that $\text{SmPt}_2\text{Cd}_{20}$ is regarded as a rare cubic system that is located in the vicinity of a FM quantum critical point.

5. Fermi surface investigation on SmIr_2Si_2

For comparison, we have extended our study into non-cubic systems, focusing on tetragonal 1-2-2 system, where anomalous HF behaviors have been observed. We have succeeded in growing high quality single crystals of SmIr_2Si_2 and observing de Haas-van Alphen signals (dHvA). The observed Fermi surfaces (FS) are different from those of LaRh_2Si_2 , suggesting a folding of the Brillouin zone caused by antiferromagnetic (AFM) ordering at 37 K. The cyclotron effective mass has been found to be ranging from 0.53 to 2.1 m_0 depending on branches. This first observation of FS properties may give a clue to understand the possible formation of HF states in AFM phases in $\text{Sm}T_2\text{Si}_2$ compounds (T : transition metals).

The abovementioned findings have clarified that Kondo behaviors observed in Sm-based intermetallic compounds are quite different from those in Ce-based HF compounds; for instance, Sm ions are in intermediate valence states despite the low Kondo temperature. These findings are expected to be keys to reveal the origin of the magnetic-field-insensitive Kondo effect in these Sm-based HF compounds.