学位論文要約（博士（理学））

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論文題名：Unusual Magnetic-Field-Insensitive Heavy Fermion States in SmT_2Al$_{20}$ (T: Transition Metals)
邦題:SmT_2Al$_{20}$ (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）

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 $YbRh_2Si_2$ and β-$YbAlB_4$, quadrupolar Kondo effect and associated NFL behaviors in PrTr_2X_{20} (*Tr*: 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 $SmOs_4Sb_{12}$ and SmTr_2Al$_{20}$ (*Tr*: 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 SmTr_2Al$_{20}$ systems. We have grown single crystals of SmTr_2Al$_{20}$ 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 $SmTa_2Al_{20}$

We have succeeded in growing single crystals of $SmTa_2Al_{20}$ 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 $\gamma \cong 3$ J/mol K^2, largest among Sm-based intermetallic compounds. Specific heat data suggest that the CEF ground state of the 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 μ_B/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 4*f* 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 $Sm_xLa_{1-x}Ta_2Al_{20}$

The La-substitution effect on the transport properties has been examined using single crystals of $Sm_xLa_{1-x}Ta_2Al_{20}$ (x = 0.01, 0.05, 0.25, 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 $Sm_xLa_{1-x}Ta_2Al_{20}$ 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 $LaTr_2Al_{20}$ (*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 $LaTr_2Al_{20}$ (*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, 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 $Ga_{0.2}V_2Al_{20}$, $Al_{0.3}V_2Al_{20}$, ScV_2Al_{20}, and LuV_2Al_{20}. The large T_c distribution in the four $LaTr_2Al_{20}$ 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 $SmPt_2Cd_{20}$

In $SmTr_2X_{20}$ compounds (*Tr*: transition metals, *X*: Zn and Cd), the 4*f* 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 $SmPt_2Cd_{20}$, a new member of $SmTr_2X_{20}$ family. It has been found that $SmPt_2Cd_{20}$ exhibits a ferromagnetic (FM) transition at T_{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 $SmPt_2Cd_{20}$ 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 $SmTr_2Si_2$ compounds (*Tr*: 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.