論文の要旨
(Thesis Summary)

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論文題目

Evolution of magnetic order in the frustrated quasikagome Kondo lattice $CeRh_{1-x}Pd_xSn$
(フラストレートした擬カゴメ近藤格子 $CeRh_{1-x}Pd_xSn$ における磁気秩序の発達)

The magnetic frustration has long been studied in insulating antiferromagnets with a triangular lattice in which all the three spins can't be antiparallel due to the antiferromagnetic (AFM) interaction. The highly correlated spin fluctuation down to zero temperature leads to the formation of a spin liquid state. In metallic Ce and Yb compounds, on the other hand, the appearance of long-range magnetic order has been thought to depend on the competition between the on-site Kondo interaction and the intersite RKKY interaction. Recently, Kondo lattice systems with Ce and Yb ions in a geometrically frustrated arrangement attract much attention. For example, Kondo lattice compounds with kagome-like geometry are expected to have an unconventional quantum critical point (QCP), which is caused by the interplay among Kondo effect, RKKY-interaction, and magnetic frustration. Thereby, an additional tuning parameter Q, magnetic frustration, has been introduced to describe the unconventional quantum critical behaviors. In a system with the spin liquid ground state, a long-range AFM order may be induced by either destruction of the symmetry of frustrated structure or the breakdown of Kondo screening.

In this thesis, we focus on CeRhSn crystallizing in the hexagonal ZrNiAl-type structure with a quasikagome lattice of Ce atoms in the basal c plane. The strong hybridization between the Ce 4f state and the 4d band from Rh2 atoms leads to a high Kondo temperature T_K □ 200 K. A zero-field QCP has been suggested by divergences in the specific heat coefficient C/T, magnetic susceptibility χ, and magnetic Grüneisen parameter. The anisotropic thermal expansion displays the critical behavior only along the a axis. The spin-flop crossover in the field dependence of C/T appears under the magnetic field of $B \parallel a = 3.5$ T. These experimental facts suggested that CeRhSn is located very close to a QCP due to the geometrical frustration.

The present work is aimed to clarify whether or not the quantum critical behavior in CeRhSn is driven by the geometrical frustration in the quasikagome lattice. For this purpose, we have prepared polycrystalline samples of $CeRh_{1-x}Pd_xSn$ and measured the electrical resistivity ρ, magnetization M, magnetic susceptibilities χ_{dc} and χ_{ac}, and specific heat C. With the increase in x from 0 to 0.75, the

hexagonal a parameter linearly increases while the c parameter deviates from the Vegard's law. The polycrystalline samples are composed of grains preferentially oriented along the c axis. Along this direction, the electric current I and magnetic field B were applied in the measurements of resistivity and magnetization, respectively.

With Pd substitution, the change in the 4f state from a delocalized state to a localized one manifests itself in $\chi(T)$ and isothermal $M(B)$. For $x \geq 0.65$, the effective moment μ_{eff} increases nearly to the value for a free Ce^{+3} ion. For $x = 0.75$, $\chi(T)$ exhibits a peak at 3 K, suggesting an AFM order. The AFM order is corroborated by a spin-flop like behaviour in $M(B)$, which reaches a saturated value of 1.4 μ_B/Ce at 5 T. Emergence of a magnetic order in $CeRh_{1-x}Pd_xSn$ for $x = 0.1$ has been observed in temperature dependence of C/T. It exhibits $-\log T$ dependence which is followed by a maximum at 0.1 K. As x increases from 0.2 to 0.4, the temperature at the maximum of C/T gradually increases to 0.7 K. It is noteworthy that the maximum temperature for each x agrees with the maximum temperature of $\chi_{\mathrm{ac}}(T)$. The magnetic entropy $S(T)$ curves for $x = 0.65$ and 0.75 are saturated to $R\ln 2$ at 10 K, confirming the doublet ground state of the localized 4f state under the crystal field.

Single crystals with $x = 0$ and 0.1 were prepared by the Czochralski method. The different ground states for $x = 0$ and 0.1 were confirmed by the measurements of temperature- and magnetic-field dependences of $\chi_{\mathrm{dc}}(T, B)$, $\chi_{\mathrm{ac}}(T, B)$, and electrical resistivity $\rho(T, B)$. The data of $\chi_{\mathrm{ac}}(T)$ with $x = 0.1$ shows a maximum at 0.15 K, close to the temperature at maximum in $C(T)$. The field dependence of $\chi_{\mathrm{ac}}(B \parallel a)$ for $x = 0$ exhibits a metamagnetic crossover at 3.5 T in agreement with the field dependence of C/T, which indicates the destruction of frustration among Ce moments in the c plane. For $x = 0.1$, the large anisotropy of $\rho(T)$, $\rho(I \parallel a) >> \rho(I \parallel c)$, is maintained. Both $\rho(I \parallel a)$ and $\rho(I \parallel c)$ show a continuous increase on cooling, which is attributed to the loss of coherent scattering. The large anisotropy $\chi_c(T) >> \chi_a(T)$ for $x = 0$ is maintained for $x = 0.1$. The positive magnetoresistance (MR) for $x = 0$ is the characteristic of coherent Kondo lattice compounds. On the contrary, MRs for $x = 0.1$ are all negative, which is the behavior of incoherent Kondo lattice. The absolute value for $I \parallel B \parallel c$ at 8 T is approximately 5 times larger than that for $I \parallel B \parallel a$, which is consistent with that of $M(B \parallel c) \cong 5M(B \parallel a)$. The magnetic and transport properties for the single crystal with $x = 0.1$ show that the magnetic frustration and Kondo effect for $x = 0$ are both suppressed. As a result, an AFM order is induced at $x = 0.1$, leaving the zero-field quantum critical point at $x = 0$.

In addition to the geometrical origin, the correlated electron systems with well separated one-dimensional spin chains are expected to exhibit the magnetic frustration driven by a delicate balance between n-th nearest neighbor interactions along the chain. Therefore, the ground state of metallic Ce-based compounds with one-dimensional Ce chains deserves to be explored.

Another purpose of this work is to study the effect of short-range correlations in the systems with a chain of Ce atoms. For this purpose, we focus on a new compound $CePd_3Sn_2$ that discovered by the present work. The single crystal x-ray diffraction analysis revealed that it crystallizes in the

orthorhombic structure with one-dimensional chains of the shortest Ce-Ce distance along the *b* axis. The measurements of C, χ, M, and ρ on polycrystalline samples were performed to explore the ground state properties of $CePd_3Sn_2$.

The $\rho(T)$ data show a metallic behavior with an obvious drop at 0.6 K, indicating a magnetic phase transition. The magnetic contribution ρ_m does not exhibit any sign of Kondo scattering. A maximum in $\chi(T)$ at 0.6 K suggests an AFM order. The isothermal magnetization $M(B)$ at 0.5 K bends at around 4 T and reaches 1.2 μ_B/Ce at 10 T. A sharp peak at 0.6 K in $C(T)$ agrees with that of the peak in $d\rho/dT$. The inelastic neutron scattering experiments revealed two excited crystal-filed doublets at 27 K and 191 K above the ground state doublet, respectively, whose energies agree with those obtained from the specific-heat measurements. Because no Kondo effect was observed in ρ_m(T), we attribute the tail from 0.6 K up to 3 K in $C(T)$ to AFM short-range correlations inherent in one-dimensional systems.