論文の内容の要旨

Low-temperature magnetization measurements of the Yb-based metamagnetic compounds

(メタ磁性を示すYb系化合物の低温磁化測定)

氏　名　　志　村　　恭　通

Heavy fermion state and quantum phase transitions in Yb-based compounds have recently attracted much attention. Since Yb^{3+} ion ($4f^{13}$) is the hole counterpart to Ce^{3+} ($4f^{1}$), formation of the heavy fermion state due to the conventional Kondo effect can be expected. There are, however, some important differences between Ce- and Yb-based compounds. Yb-based compounds tend to exhibit a valence transition/crossover. Indeed, several Yb-based heavy fermion compounds are considered to be close to a quantum critical point of the valence transition. Another important feature of Yb compounds is that Yb^{3+} ions have a larger angular momentum $J = 7/2$ than $J = 5/2$ in Ce^{3+}. This large J value often results in a large (pseudo) degeneracy, especially in a cubic environment. By contrast, under a strong CEF of lower symmetry, the large J value would result in a strong magnetic anisotropy. These features of Yb-based compounds would lead to a rich variety of the quantum criticality and quantum phase transitions, of which not much has been known. In the present thesis, we studied field-induced phase transitions on single crystals of some ytterbium (Yb)-based compounds at very-low temperatures down to ~ 0.1 K by means of DC magnetization measurements. These include $YbCo_2Zn_{20}$, $YbIr_2Zn_{20}$ and Yb_2Pt_2Pb, all of which indicate “metamagnetic” behavior, a step-like increase in the field dependence of the magnetization $M(H)$.

The cubic heavy-fermion compounds YbT_2Zn_{20} (T = Fe, Co, Ru, Rh, Os and Ir) are known to exhibit no long-range order at zero field and in ambient pressure. In these compounds, the Yb ion is located in a cage formed by sixteen Zn ions. Among the YbT_2Zn_{20} family, $YbCo_2Zn_{20}$ is particularly of interest because it exhibits a gigantic electronic specific heat coefficient of 8000 mJ/molK^2. In the previous

AC susceptibility measurements, metamagnetism that can be ascribed to the heavy fermion state has been observed along the [100] direction with the critical field of $\mu_0 H_m = 0.6$ T at low temperatures below ~ 0.3 K. We confirmed by the DC magnetization measurements that the metamagnetic behavior is nearly isotropic reflecting the cubic symmetry, in contrast with other heavy-fermion systems with lower symmetry in which metamagnetism appears only for a particular field direction. In the course of the experiment, we found a new metamagnetic transition above 6 T only for $H//[111]$. For the same direction, a clear kink has been observed in the temperature dependence of the magnetization $M(T)$ above 6 T. These results indicate the existence of a new field-induced ordered phase whose order parameter is probably an quadrupole moment. This field-induced phase transition can be explained by a crossing in the pseudo-sextet CEF state formed by doublet and quartet for Yb^{3+}. A strong cubic anisotropy of $M_{[100]} > M_{[110]} > M_{[111]}$ observed above 2 T can be also understood by the CEF scheme. Thus, our magnetization data confirm that the heavy fermion state in $YbCo_2Zn_{20}$ is arising from the pseudo-sextet state. These results also suggest the stability of the Yb^{3+} state in high magnetic fields, possibly due to a suppression of the heavy-fermion state by a magnetic field.

$YbCo_2Zn_{20}$ is also known to undergo a quantum phase transition of a magnetic ordering by applying pressure at zero field. We attempted to drive the quantum phase transition by doping Ni for Co. We have grown the single crystals of $Yb(Co,Ni)_2Zn_{20}$ (Ni/Co ~ 0.013) by the Zn self-flux method. While no evidence of a magnetic transition has been obtained, the metamagnetic behavior at 0.6 T disappears by the small Ni doping. Interestingly, the temperature dependence of the magnetic susceptibility $\chi(T)$ in $Yb(Co,Ni)_2Zn_{20}$ continues to increase even below 100 mK, in contrast to the behavior of $\chi(T)$ in $YbCo_2Zn_{20}$, which becomes constant below ~ 0.3 K. The results strongly suggest that the Ni doping makes $YbCo_2Zn_{20}$ to approach the quantum critical point.

$M(H)$ in $YbIr_2Zn_{20}$ has been reported to exhibit anisotropic metamagnetism at $\mu_0 H_m = 9.7$ T, 12 T and 13.4 T for $H//[100]$, [110] and [111], respectively. This metamagnetic behavior is also not due to a long-range ordering. We derived the temperature dependence of the differential susceptibility $\mathrm{d}M/\mathrm{d}H$ at the critical field H_m of metamagnetism from the $M(H)$ data at several temperatures below 4.2 K. $\chi_m(T) \equiv (\mathrm{d}M/\mathrm{d}H)_{H=H_m}$ indicates a weak upturn on cooling and does not saturate down to the base temperature of 0.09 K. This behavior is at odds with the typical Fermi liquid behavior. We confirmed that the upturn is not due to a Yb nuclear magnetization by carefully evaluating the effect of a hyperfine coupling between nuclear spins and the $4f$ magnetic moment. This upturn in $\chi_m(T)$ provides possible evidence of the quantum criticality of the Yb valence crossover.

By contrast, $4f$ electrons in the tetragonal Yb_2Pt_2Pb are well localized and provide local moment magnetism. Magnetic properties of Yb_2Pt_2Pb can be well explained by the CEF model for Yb^{3+}. Yb^{3+} ions in Yb_2Pt_2Pb are arranged in a two dimensional network formed by mixed rectangles and isosceles triangles, topologically equivalent with the Shastry-Sutherland lattice. Because of the strong CEF of low local symmetry, the magnetic moments of Yb ions show strong Ising type anisotropy with the easy axis pointing along either [110] or [1$\bar{1}$0] direction in the two-dimentional plane of the Shastry-Sutherland lattice. Yb_2Pt_2Pb is known to exhibit an antiferromagnetic ordered phase (phase I) below $T_N = 2.1$ K and a field-induced ordered phase (phase II) above ~ 1 T. The anisotropic magnetic phase diagram as well as the in-plane anisotropy in $M(H)$ have been explained by Ochiai *et al.* on the basis of an Ising spin model with the orthogonal Ising axes along [110] or [1$\bar{1}$0]. According to the model, the CEF ground state wave function is predominantly composed of $|J_z = \pm 7/2\rangle$, where the quantization z-axis is either [110] or [1$\bar{1}$0]. We studied the field-induced phase transitions in Yb_2Pt_2Pb at very low temperatures. Cascade metamagnetic transitions are observed around the I-II phase boundary at low temperatures. Interestingly, a nearly linear increase in $M(H)$ has been observed in phase II even at a very low temperature of 0.08 K. In addition, the phase transition from phase II to the paramagnetic state is found to be of second order, reminiscent of a spin-flop state in ordinal antiferromagnets. However, the appearance of a spin-flop state is highly incompatible with the Ising nature of the Yb moment. We show that this discrepancy can be solved by introducing a high-rank multipole interaction. The CEF ground state wave function, predominantly composed of $|J_z = \pm 7/2\rangle$, can be represented by a pseudospin whose z component is J_z whereas x and y components are high-rank multipole moments. Then, a spin-flop-like state can be realized in a magnetic field parallel to the z axis even in the presence of a strong Ising anisotropy of the magnetic moment. Namely, phase II can be regarded as a canted state of the pseudospins. In the simplest approximation, the relevant high-rank multipole component of the pseudospin is a rank-7 octacosahectapole (128-pole) moment. We have done numerical calculations based on a mean-field model incorporating the interaction between the rank-7 multipole moments. The basic features of the magnetic phase diagram as well as the magnetization $M(H, T)$ are successfully explained by the model.