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Metamagnetic-Like Anomalies in f-Electron Systems Investigated by Specific Heat Measurements

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We report the results on the specific heat (C) and magnetocaloric effect measurements in 4f- and 5f- compounds exhibiting metamagnetic-like anomaly (MLA) at field (H) around H_M from the paramagnetic ground state. The MLA in Ce-compounds ($CeRu_2Si_2$ and $CeCu_6$) and UCoAl have both close resemblance and apparent difference; the first order character has been confirmed for UCoAl but not for the Ce compounds. For the Ce compounds, a single peak $C(H)/T$ at H_M at lowest temperature splits into double peaks at finite temperatures. For UCoAl, a step-like decreases of $C(H)/T$ across H_M at lowest temperature changes to a peak structure around HM at higher temperatures.

KEYWORDS: metamagnetic transition, $CeRu_2Si_2$, $CeCu_6$, UCoAl, specific heat, magneto-caloric effect

§1. Introduction

The metamagnetic-like increase of magnetization (M) from a paramagnetic ground state observed in Ce compounds, such as $CeRu_2Si_2$,[1–3] $CeCu_6$,[4] $CeNi_2Ge_2$,[5] $CeFe_2Ge_2$,[6] is thought to be one of the most attractive subjects in highly correlated electron systems. For $CeRu_2Si_2$, which is the most intensively investigated material, there remains an interesting puzzle. De Haas-van Alphen experiment[7] suggests a clear change of Fermi surface (FS) across H_M ($\sim$ 7.7T), suggesting the change of f-electron character from intinerant to localized. On the other hand, the low temperature M measurement has ruled out the possibility of 1st order phase transition at H_M.[8] No sudden change of FS was inferred also from the transport measurements.[9] Another puzzle is the strange two phase boundaries, facing each other across H_M, in the $H-T$ phase diagram determined from the thermal expansion under magnetic fields,[10] which has not been confirmed by the other physical properties.

The specific heat (C) measurement at low T is a powerful tool to investigate the ground state of magnetic systems. However, the capability has not been fully utilized for the MLA, since C is usually measured as a function of T under discrete steps of constant H. We have designed a new technique to measure the field dependence of C quasi-continuously.[11]

We first summarize a new feature found in the H-dependence of C in $CeRu_2Si_2$, which is discussed in correlation with the fine structure in the quasi-particle density of states (DOS) near the Fermi energy (ϵ_F). We then briefly shows our preliminary result on $CeCu_6$, which also exhibits MLA near 1.7T. In order to understand such MLA in Ce compounds, the comparison with MLA in 5f- electron system is helpful. We have thus investigated the H-dependence of C also in a paramagnetic compound UCoAl, which was recently reported to exhibit MLA at around 1T.[11]

§2. Experiment

Single crystals of $CeRu_2Si_2$, $CeCu_6$ were grown by Czochralski pulling method using a tetra-arc furnace or a RF furnace. UCoAl single crystal was grown by the mineralization method. The specific heat was measured down to 100 mK by a semi-adiabatic heat-pulse method using a dilution refrigerator equipped with a 8 Tesla superconducting magnet. Using the newly designed system, the field dependence of specific heat can be measured under a quasi-isothermal condition.

§3. Results and Discussion

For $CeRu_2Si_2$, we first measured the ordinary T-dependence of C in several values of H including the H_M for comaprison with the previous report as shown in Fig.1. In the T-range, the lattice specific heat contribution $C_L = \beta T^3$ (β=0.295mJ/K^{4}mol) estimated from $LaRu_2Si_2$ is negligible. In zero field, C/T becomes almost constant ($\gamma \sim$ 370mJ/K^{2}mol) below $\sim$2K, which is consistent with the reported data.[3] The temperature where C/T starts to saturate decreases with increasing field up to H_M, where it does not saturate down to 0.2K. Above H_M, C/T again shows a saturation behavior, thereby C/T at 0.2K shows a maximum as a function of H. On the other hand, C/T at high T shows a minimum near H_M. To clarify such a delicate change of C/T with H, we have directly measured the H-dependence of C/T.

The results at selected values of T are shown in Fig. 2.

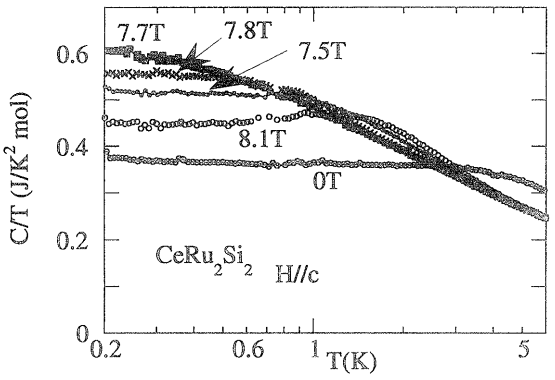

Fig. 1. Temperature dependence of C/T under selected values of magnetic field for $H//c$.

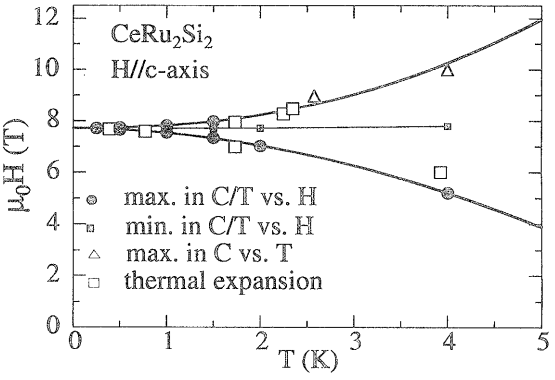

Fig. 3. $H-T$ phase diagram for the anomalies in the specific heat and the thermal expansion measurements.

At 0.25K, C/T shows a sharp single peak centered at $H_{\rm M}$. Sakakibara *et al.*[8] has predicted such a peak by analyzing the T-dependence of M based on the thermodynamic Maxwell relation. It should be noted that there exists no hysteresis within the experimental accuracy. The most prominent feature in Fig.2 is the double peak structure. The distance between the two peaks becomes wider with increasing T. At 4.0K, the peak can be only barely visible. The peak and minimum positions

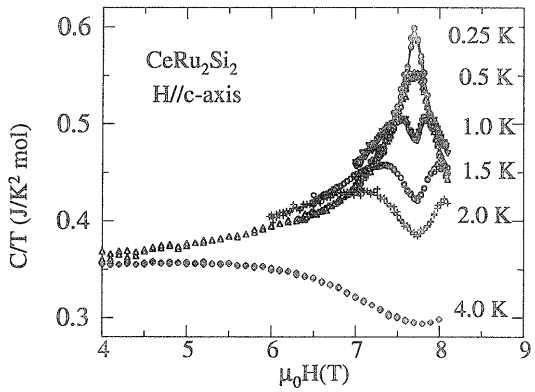

Fig. 2. Field dependence of C/T for several values of temperature.

are plotted as a $H-T$ phase diagram in Fig.3 along with the reported anomaly in the thermal expansion.[10] The positions approximately agree in the two experiments, which suggests that the inherent mechanisms might be same.

A sharp peak in the-partial-density of states in the hybridized-band $\rho_{\rm M}(\epsilon)$ slightly above $\epsilon_{\rm F}$ at H=0 was theoretically predicted as an origin of the metamagnetic behavior.[11,12] A peak in C_V/T versus H at 0K was explained in the same model; C/T shows a peak when the main DOS peak is shifted to $\epsilon_{\rm F}$ due to Zeeman splitting, while the H-dependence of C/T ($\sim |H-H_{\rm M}|^{-1/2}$) at low T reflects the energy dependence of DOS(ϵ) $\sim (\epsilon-\epsilon_0)^{-1/2}$ with $\epsilon_0 = \mu_{\rm eff} H_{\rm M}$.

Based on the same model, we try to reproduce the double-peak structure of $C/T-H$ curves as shown in Fig.4. C/T at finite T is represented as,

$$C/T = 2\int dx\{\rho_{\rm M}[2Tx - \mu_{\rm eff}(H_{\rm M}-H)] + \rho_{\rm M}[2Tx - \mu_{\rm eff}(H_{\rm M}+H)]\}x^2 sech^2(x),$$

where the 1st and 2nd terms represent the contributions from the up-spin and down-spin bands, respectively. The thermal weighing factor $(\epsilon/k_{\rm B}T)^2 sech^2(\epsilon/k_{\rm B}T)$, which has two maxima above and below $\epsilon_{\rm F}$, is essential. At 0K, when the DOS peak just passes $\epsilon_{\rm F}$ with increasing H, C/T exhibits a peak. At finite T, C/T is enhanced when the peak position coincides with one of the $x^2 sech^2(x)$ peaks, which leads to the double peak structure. For comparison with C_V, the volume effect must be taken into account. According to the precise investigation of the thermal expansion,[13] the volume effect causes no fundamental change to the metamagnetic anomaly, though it somewhat changes the magnitude and width of anomaly. At this stage, we may say that the peak in DOS near $\epsilon_{\rm F}$ is one of the possible origins of MLA in $CeRu_2Si_2$.

For comparison with the above results, $CeCu_6$ is only one possible Ce-compound on which the effect of metamagnetic behavior is observable in the specific heat within a reasonable field range. Fig.5 (a) shows the field dependence of C/T on $CeCu_6$ for $H//c$.[11]

At 0.2K, there exists a bump near 1.7T where MLA was reported. At 0.5K, the peak is split into two bumps as shown by the arrows. In the magneto-caloric experiment, such structures related with MLA could be more clearly observed as shown in Fig.5 (b). All these features resemble with those in $CeRu_2Si_2$, though they are less clear in $CeCu_6$, which is natural since the structure in M is also weaker in $CeCu_6$.[4] This fact suggests that the MLA in both compounds has similar origin.

Usually, the itinerary of 5f electrons is thought to be in between 4f and 3d electrons. The mechanism of the

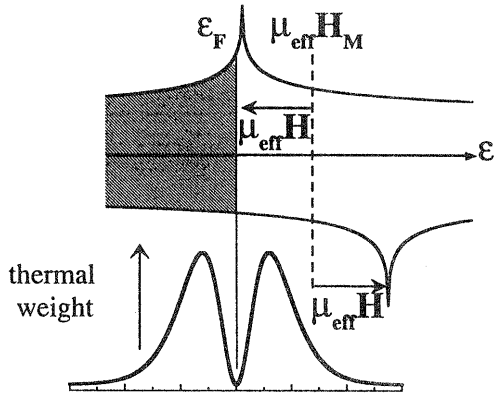

Fig. 4. Schematic figure of the density of states and the thermal weighing factor for the specific heat.

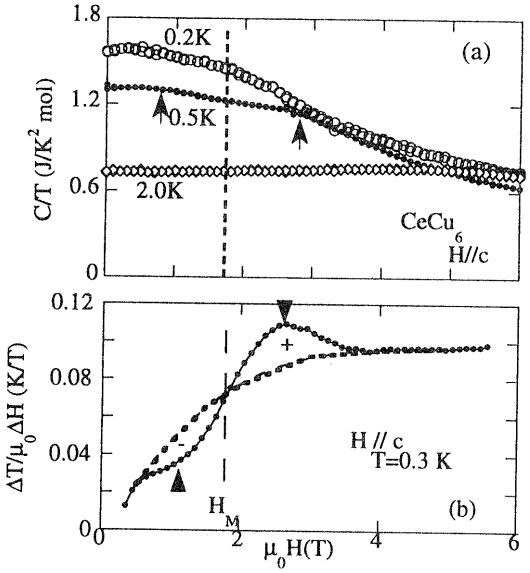

Fig. 5. Field dependences of (a) specific heat and (b) magnetocaloric effect for $CeCu_6$.

metamagnetic transition in typical 3d metamagnet, such as YCo_2 and $LuCo_2$ is ascibed to the itinerant-electron metamagnetism resulting from the high 3d-band DOS near ϵ_F.[14] It is of interest to compare the effects of MLA on specific heat between UCoAl and $CeRu_2Si_2$.

Fig.6 shows, the comparison of the field dependence of C/T and M for UCoAl in the same geometry. We observed a clear hysteresis behavior both in C/T and M at least below 3K, which indicates the transition to be 1-st order. C/T shows a clear decrease across H_M at low temperatures, reflecting the change in DOS. Taking into account the transport results,[15] we infer that the MTA in UCoAl is the 5f-band split metamagnetism resembles with those found in 3d-system.

§4. Conclusions

The MLA in $CeRu_2Si_2$ and $CeCu_6$ shows close similarities: the absence of hysteresis rules out the 1st order nature of the transition, C/T exhibits a double peak structure against H, etc. and are ascribed to the fine

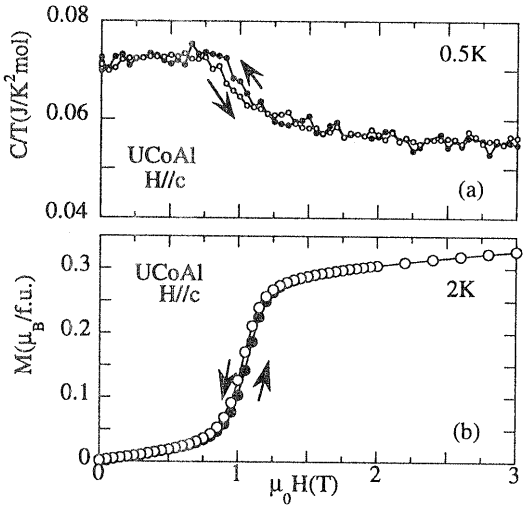

Fig. 6. Field dependence of (a) C/T and (b) M for UCoAl.

structure in the partial DOS near ϵ_F. The metamagnetic transition in UCoAl is of 1st order, and the high field state could be described as 5f-band split ferromagnetic state.

Acknowledgements

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