MAGNETIC PROPERTIES OF THE AuCu3-TYPE Sm-COMPOUNDS

B. Liu, M. Kasaya, T. Kasuya

JOURNAL DE PHYSIQUE
Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

MAGNETIC PROPERTIES OF THE $AuCu_3$-TYPE Sm-COMPOUNDS

B. Liu, M. Kasaya and T. Kasuya

Department of Physics, Faculty of Science, Tohoku University, Sendai 980, Japan

Abstract. – Resistivity, susceptibility, specific heat and magnetization measurements on single crystals $SmPb_3$, $SmPd_3$ and $SmTl_3$ are performed. The phase transition of the antiferromagnet, $SmPb_3$, is of first order at 5.1 K. $SmPd_3$ shows an antiferromagnetic ordering at 1.36 K. $SmTl_3$ behaves as a ferromagnet with T_c =8.6 K. The ground states of Sm^{3+} (J= 5/2) ions in these Sm-compounds are Γ_8 quartets.

Recently, study of the valence fluctuation in the 4f electron system has been rapidly growing. In particular, many cerium compounds, having simple configuration of $4f^0$ and $4f^1$, exhibit anomalous physical properties which can be considered as the characteristic of Kondo lattice. In contrast, the Kondo systems containing rare earth with many 4f electron(hole) of shells are exceedingly rare and, as a rule, poorly documented. Considering the same value of $J = 5/2$ in Ce^{3+} and Sm^{3+} ions, it is worthwhile to perform experiments on Sm-compounds and compare with the isomorphous Ce-compounds systematically in order to clarify the effects of Crystal-Electric Field (CEF) on the ground state properties of Kondo ions.

It is well known that both CeX_3 and SmX_3 (X = Pd, In, Sn, Pb, Tl) have the cubic $AuCu_3$ structure [1]. There have been many detailed studies on magnetic and transport properties of the $AuCu_3$ structure Ce-compounds except $CeTl_3$. The series of CeX_3 (X = Pd, In, Sn, Pb) show various interesting characteristics of dense Kondo behaviour. On the other hand, only a few studies have been done on the SmX_3 (X = Pd, In, Sn, Pb). The magnetic susceptibilities of SmX_3 (X = Pd, In, Sn, Pb) have been reported by several authors and are believed to be antiferromagnets with T_N =16 K, 11 K and 6 K for $SmIn_3$, $SmSn_3$ and $SmPb_3$, respectively [2, 3, 4]. The magnetic phase transition has not yet been observed on $SmPd_3$ at low temperature range, namely down to 2 K [5]. However, there was not any experimental study of $SmTl_3$ and $CeTl_3$. It is previously reported by us that $SmSn_3$ and $SmIn_3$ are antiferromagnets accompanied with the quadrupolar orderings and the ground states of Sm^{3+} ions are Γ_8 quartets [6]. Furthermore, as the first examples in Sm-compounds, $SmSn_3$ and $SmIn_3$ show a kind of dense Kondo behaviour. In this paper, we will present the experimental results on $SmPb_3$, $SmPd_3$ and $SmTl_3$ and compare them with those of $SmSn_3$ and $SmIn_3$. Here, all the samples used were single crystals prepared in the sealed Mo crucible by the Bridgman method.

The results of specific heat measurements of $SmPb_3$, $SmPd_3$ and $SmTl_3$ in the temperature range of 0.6 K to 20 K are shown in figure 1. The very sharp λ-type anomaly for $SmPb_3$ indicates that the magnetic phase transition is of first order. The peak observed

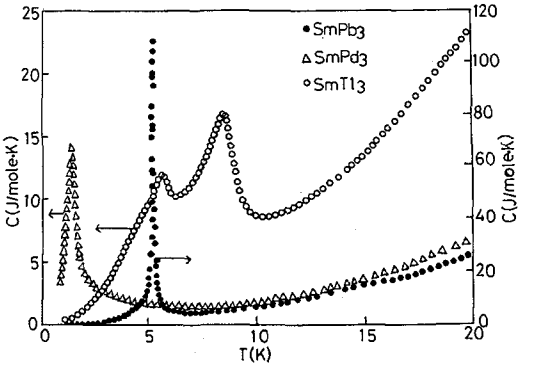

Fig. 1. – The specific heats C *vs.* temperature T for $SmPb_3$, $SmPd_3$ and $SmTl_3$.

for $SmPd_3$ at 1.36 K correspond to the magnetic phase transition. For $SmTl_3$, two peaks in the specific heat occur at 8.6 K and 5.4 K. The former peak corresponds to Curie temperature. At present, the origin of the latter peak is not clear. The specific heats at lowest temperature show the Fermi liquid-like T-linear behaviours with the large γ-values, $\gamma \approx 100$, 280 and 200 $mJ/mole * K^2$ for $SmPb_3$, $SmPd_3$ and $SmTl_3$, which have similar values to those of $SmSn_3$ and $SmIn_3$. To compare the specific heat of $SmTl_3$ with the isomorphous Ce-compounds, we also measured the specific heat of $CeTl_3$ and found a peak at T = 3.4 K. The magnetic entropies are obtained by subtracting the specific heats of $LaPb_3$, $LaPd_3$ and $LaTl_3$ from those of $SmPb_3$, $SmPd_3$ and $SmTl_3$. Above their magnetic phase transitions, the entropies for $SmPb_3$, $SmPd_3$ and $SmTl_3$ increase nearly up to R ln 4, are the same as those of $SmSn_3$ and $SmIn_3$. The temperature dependence of the entropies suggest that the ground states of CEF splitting $J = 5/2$ level are Γ_8 quartets. However, the Γ_8 quartets ground levels are most clearly shown by the strong Curie-like softening of the ultrasonic velocities, or the elastic constants of $SmPb_3$, $SmPd_3$ and $SmTl_3$ [7]. On the other hand, the entropy of $CeTl_3$ only reaches nearly R ln 2 up to 10 K in agreement with a Γ_7 (doublet) ground state. Indeed, it was known that the $AuCu_3$-type Ce-compounds, i.e., $CePb_3$ $CePd_3$, $CeIn_3$ and $CeSn_3$, are Γ_7 ground states in the cubic crystal field.

The temperature dependences of the magnetic susceptibilities are measured between 2.0 K and 300 K. We have analysed the susceptibility data of $SmPb_3$, $SmPd_3$ and $SmTl_3$ (corrected for non 4f contribution, individually) in order to dertermine the ground states and to estimate the CEF splittings of Sm^{3+} ions. The expression for 4f susceptibility in the presence of exchange and excited multiplets has been given by De Wijn *et al.* to be

$$\chi_{4f} / N\beta = \sum_J p(J) \left[\frac{a}{T} + b\right] - \\ - \frac{2J_{ff}}{\beta} \left\{ \sum_J p(J) \left[\frac{(g_f - 1)\, a}{g_f T} + b \right] \right\} \\ \times \left\{ 1 - \frac{2J_{ff}}{\beta} \sum_J p(J) \left[\frac{(g_f - 1)^2\, a}{g_f^2 T} + b \right] \right\}^{-1} \quad (1)$$

where the symbols have the same meaning as in reference [8].

In the first approximation the effect of the cubic crystal field acting on Sm^{3+} can be included by multiplying the first T^{-1} term in the J summation of both numerator and denominator of equation (1) by the factor [9]

$$f(T) \left[5 + 26e^{-x} + \right. \\ \left. + (32/x)\left(1 - e^{-x}\right)\right] / 21 \left(1 + 2e^{-x}\right) \quad (2)$$

where $x = W / KT$ and W is the energy separation between the doublet and the quartet energy levels, and has a sign which is positive if the doublet lies lowest and negative if the quartet lies lowest. Figure 2 shows the best fits to χ_{4f} obtained using the following parameters: $J_{ff} = -33 \pm 4$, -2 ± 1, 6 ± 2 and $W = -60 \pm 5$, -50 ± 5, -150 ± 10 in Kelvins for $SmPb_3$, $SmPd_3$ and $SmTl_3$. The negative values of J_{ff} for $SmPb_3$ and $SmPd_3$ indicate that the phase transitions are antiferromagnetic orderings. The magnitudes of CEF splitting obtained from susceptibilities are similar to those analyzed from the ultrasonic velocities [7]. Furthermore, the $\Gamma_8 - \Gamma_7$ splitting of $SmPb_3$ is about 60 K in full agreement with the variation of CEF parameters throughout the RPb_3 series [10]. Such variations of CEF parameters are also observed for $SmSn_3$ and $SmIn_3$ throughout the RSn_3 and RIn_3 series. It is to be noted that the saturation magnetic moment of only $\approx 0.08\ \mu_B$ is very small in comparison with the value expected for the Γ_8 ground state of Sm^{3+} ions. The reduction of the magnetic moment residing on the samarium is not clear at present but seems due to the Kondo Effect.

Finally, the electrical resistivities of $SmPb_3$, $SmPd_3$ and $SmTl_3$ are reported. The anomalies observed for $SmPb_3$ and $SmTl_3$ at 5.1 K and 8.6 K correspond to the magnetic phase transitions, respectively. However, the Kondo behaviours are not clearly observed like in $SmSn_3$. The temperature dependence of the resistivity

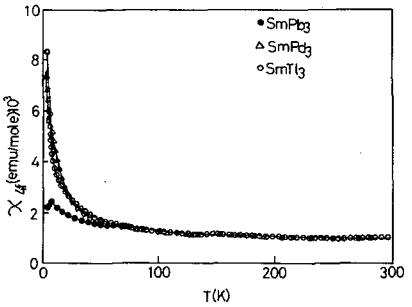

Fig. 2. – The temperature dependence of magentic susceptibilities (corrected for non 4f contributions) for $SmPb_3$ (•), $SmPd_3$ (△) and $SmTl_3$ (○). The full lines are the best fits to χ_{4f} of $SmPb_3$, $SmPd_3$ and $SmTl_3$.

is not so impressive and this may be dominated by the Kondo scattering mechanism for samarium compounds given by us [11].

In conclusion, the magnetic properties are studies on the single crystals $SmPb_3$, $SmPd_3$ and $SmTl_3$. It is found that $SmPd_3$ shows an antiferromagnetic ordering at $T = 1.36$ K. $SmTl_3$ undergoes a ferromagnetic phase transition with $T_c = 8.6$ K. The CEF ground states for $SmPb_3$, $SmPd_3$ and $SmTl_3$ are Γ_8 quartets as in $SmSn_3$ and $SmIn_3$. Indeed, the experimental results for $SmPb_3$, $SmPd_3$ and $SmTl_3$ agree better with Γ_8 lower than Γ_7 which again agree with the prediction of the point charge model for a fourth degree term (a sixth degree term has no effect on the lowest multiplet).

[1] Landelli, A. and Palenzona, A., Handbook of the Physics and Chemistry of Rare Earth, Vol. 2, Eds. K. A. Gschneidner and L. R. Eyring (North Holland Publ. Comp., Amsterdam) 1979, p. 1.

[2] Tsuchida, T. and Wallace, W. E., *J. Chem. Phys.* **43** (1965) 3811.

[3] Buschow, K. H., De Wijn, H. W. and van Diepen, A. M., *J. Chem. Phys.* **50** (1969) 137.

[4] Lethuillier, P., Quezel-Ambruanz, G. and Percheron, A., *Solid State Commun.* **21** (1973) 105.

[5] Gardner, W. E., Penfold, J., Smith, T. F. and Harris, I. R., *J. Phys. F* **2** (1972) 133.

[6] Kasaya, M., Liu, B., Sera, M. and Kasuya, T., *J. Magn. Magn. Mater.* **52** (1985) 289.

[7] Endoh, D., Goto, T., Tamaki, A., Liu, B., Kasaya, M., Fujimura, T. and Kasuya, T., to be published.

[8] de Wijn, H. W., van Diepen, A. M. and Buschow, K. H. J., *Phys. Rev.* **161** (1967) 253.

[9] Jones, E. D., *Phys. Rev.* **180** (1969) 455.

[10] Lethuillier, P. and Chaussy, J., *J. Phys. France* **37** (1976) 123.

[11] Kasuya, T., Kasaya, M., Takegahara, K., Iga, F., Liu, B. and Kobayashi, N., *J. less-Common Met.* **127** (1987) 337.