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Anomalous Transport and Thermal Properties of $YInCu_4$

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The magnetic susceptibility, electrical resistivity and low-temperature specific heat have been measured for a ternary compound $YInCu_4$ which forms the cubic C15b-type crystal structure. $YInCu_4$ is diamagnetic, which is probably due to the low density of conduction electrons. With decreasing temperature, the electrical resistivity increases to a broad maximum around 300 K and then decreases gradually to the lowest temperature. The temperature dependence of C/T, where C is the specific heat, is convex downward over the entire temperature range of measurement to 1.5 K. $YInCu_4$ may be classified as a semimetal. The origin of the transport and thermal anomalies may be explained in the framework of semimetallic substances.

[$YInCu_4$, magnetic susceptibility, electrical resistivity, specific heat, semimetal]

With indium and copper, the heavy rare earth elements (R=Gd, Lu) form cubic compounds, $RInCu_4$. The crystal structure is of the C15b-($MgSnCu_4$-) type, where R and In atoms are in a face-centered cubic (fcc) arrangement. In a unit cell with R atoms at the corners, In atoms situate at the (1/4, 1/4, 1/4) positions. Several anomalous magnetic properties of some $RInCu_4$ compounds have been previously reported.[1-4] $YbInCu_4$ shows a first-order valence transition at $T_v=40$–50 K from the localized moment state of Yb^{3+} above T_v to the intermediate valence (Fermi liquid) state below T_v, associated with discontinuous changes in such factors as the susceptibility, unit cell volume and nuclear spin-lattice relaxation time.[1-3] Though a similarity to the α-γ transition of pure metallic cerium has been pointed out,[5] the origin is not completely understood yet. The compound with gadolinium, $GdInCu_4$, orders antiferromagnetically at a very low temperature of $T_N=6.9$ K, in spite of significant antiferromagnetic interaction expected from the Curie-Weiss behavior with the paramagnetic Curie temperature of -45 K.[4] We explained such anomalous magnetism of $GdInCu_4$ by introducing the magnetic frustration on the fcc lattice. The transport properties of $GdInCu_4$ are also anomalous. The electrical resistivity increases with decreasing temperature to 80 K, shows a broad maximum, then decreases gradually at temperatures below 80 K to a sharp peak around T_N. The high-temperature part of the resistivity exhibits $-\log T$ temperature dependence above 200 K. At the present stage, the origin of this particular temperature dependence is an open question. Such temperature dependence has been also observed in some other $RInCu_4$ compounds.[6] In order to elucidate these anomalous properties observed in the $RInCu_4$ systems, it is necessary to investigate reference materials which do not contain magnetic ions. It has been reported that the electrical resistivity of $LuInCu_4$ is normally metallic, although the specific resistivity is fairly large.[7] We found that another nonmagnetic compound, $YInCu_4$, exhibits this anomalous temperature dependence of resistivity. In this letter, we present results on the susceptibility, electrical resistivity and low-temperature specific heat of $YInCu_4$.

Polycrystalline samples of $YInCu_4$ were prepared in an argon-arc furnace from mother metals, 99.9% Y and 99.99% In and Cu. The samples were annealed at 800°C in evacuated quartz tubes for one week. Powder X-ray diffraction measurements showed the presence of (200) and (420) diffraction peaks, which discriminate the C15b structures from the C15 structure of the cubic Laves phase. The magnetic susceptibility was measured with a torsion balance magnetometer from 30 K to room temperature. Below 30 K we could not obtain reliable results because the paramagnetic contribution of the sample holder (blank) became dominant at lower temperatures, masking the very small susceptibility of samples. The electrical resistivity was measured by means of a four-probe method for several specimens cut from different ingots. The data for all ingots were nearly identical except at very low temperatures, as will be mentioned below. In this letter we show a typical result of measurement from 4.2 K to 800 K. The specific heat was measured by means of a heat pulse method in an adiabatic cell from 1.5 K to 20 K.

Figure 1 shows the temperature dependence of the magnetic susceptibility for $YInCu_4$. The susceptibility is negative over the entire temperature range of measurement. This indicates the low density of conduction electrons in this system. Such diamagnetic behavior was also observed for the isostructural nonmagnetic compound $LuInCu_4$. Takegahara and Kasuya[5] have calculated the band structure of $LuInCu_4$ and suggested that $LuInCu_4$ is a compensated semimetal with a small carrier density. Then the conduction band structure of the other $RInCu_4$ compounds may also be semimetallic. The present magnetic susceptibility is consistent with this idea.

Figure 2 shows the temperature dependence of the electrical resistivity of $YInCu_4$ from 4.2 K to 800 K. A sudden, but small, decrease of the electrical resistivity was observed below about 5 K (also see Fig. 3). It is not clear whether the anomaly is intrinsic or not. We suppose that this is caused by the superconductivity of a small amount of impurity phases such as pure indium and other intermetallic compounds precipitating in the grain boundary, because we did not detect any sign of phase transition in the specific heat measurement, as will be mentioned below. We do not discuss the low-temperature anomaly in this report. At higher temperatures, with decreasing temperature, the resistivity increases to a broad maximum between 300 K and 250 K, then decreases to the lowest temperature. The

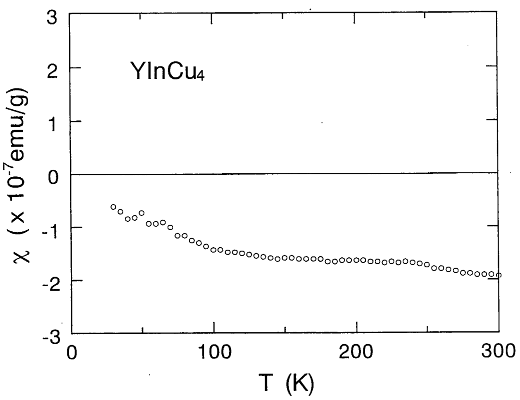

Fig. 1. The temperature dependence of the magnetic susceptibility for $YInCu_4$.

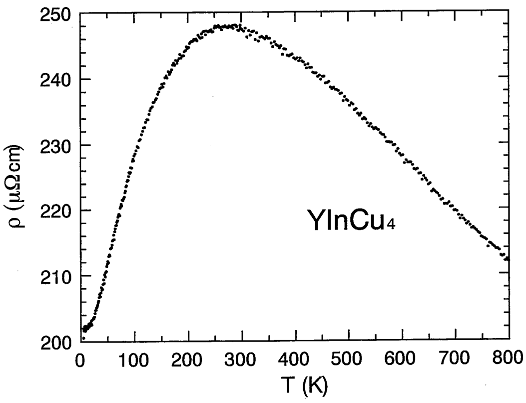

Fig. 2. The temperature dependence of the electrical resistivity for $YInCu_4$ between 4.2 K and 800 K.

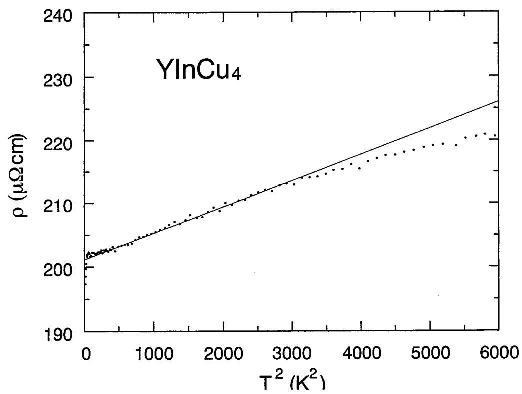

Fig. 3. The electrical resistivity of $YInCu_4$ at low temperatures as a function of the square of temperature. The straight line was derived from the equation $\rho=\rho_0+AT^2$, where $\rho_0=201\ \mu\Omega$ cm and $A=0.0042$ $\mu\Omega$ cm/K^2.

specific resistivity is comparatively large, about 200 $\mu\Omega$ cm. Similar temperature dependence was observed for $GdInCu_4$, although the temperature of the maximum, T_{max}, was lower 80 K.[4] A study of the pseudo-ternary system, $(Gd_{1-x}Lu_x)InCu_4$ indicated that the T_{max} is strongly correlated with the unit cell volume, i.e., the larger the volume, the lower the T_{max}.[8] Since the lattice parameter of $YInCu_4$ (7.211 Å) is slightly smaller than that of $GdInCu_4$ (7.235 Å), the present result also support this relation. This temperature dependence is similar in some points to that of the low-carrier heavy fermion system Yb_4As_3,[9] in which the electrical resistivity increases with temperature, reaches a maximum around 150 K and then decreases to the temperature of charge ordering at 300 K. Below 100 K, it exhibits T^2 dependence, and the coefficient of the T^2 term, A, yields the value $A/\gamma^2 = 1.0 \times 10^{-5}\ \mu\Omega$ cm (mol K/mJ)^2, where γ is the coefficient of the temperature-linear term of specific heat, which was considered to be evidence of the heavy fermion character. The dense Kondo behavior was suggested because of the negative temperature dependence of the resistivity between 150 K and 300 K. Tentatively, we plot the resistivity of $YInCu_4$ against T^2 below 77 K in Fig. 3. It seems that the resistivity varies as T^2 below 60 K. Since there are no magnetic ions in $YInCu_4$, this temperature dependence cannot be ascribed to the Fermi liquid behavior of magnetic spins. It is not clear whether the similarity to the low-carrier system is accidental or not. Plausibly, the temperature dependence may be ascribed to the crossover of the semimetallic and semiconducting behavior or to the electron-electron (hole) scattering between small Fermi surfaces as a characteristic of the compensated semimetal. The result of the band calculation of $LuInCu_4$[5] indicates the existence of small hole and electron pockets. It is plausible that the dimensions of these Fermi surfaces are sensitive to the lattice constant, and that the semimetallic characteristics are enhanced in $YInCu_4$, which has a lattice constant larger than that of $LuInCu_4$. However, no quantitative explanation of the dependence is found in the present stage.

Figure 4 shows the temperature dependence of C/T between 1.5 K and 20 K, where C is the specific heat. The monotonous change of C/T indicates no phase transition in this temperature range. The temperature derivative of C/T is positive over the entire temperature range, and no temperature-linear range of C is seen. This temperature dependence is inexplicable in the category of metallic substances, of which the specific heat consists of electronic and lattice contributions. The γ value estimated from an extrapolation of C/T to 0 K is

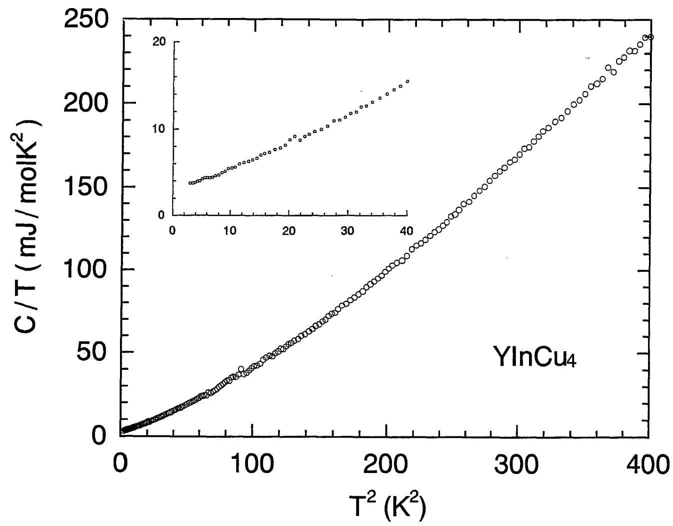

Fig. 4. The C/T vs T curve, where C is the specific heat, for $YInCu_4$. The inset shows the low-temperature part.

3-4 mJ/mol K^2.

Usually the T^2 term of electrical resistivity is observed as a many-body effect of some degree of freedom. Miyake *et al.*[10] have suggested that, in some substances with particular crystal structures such as the A15, C15 and Chevrel types, strong electron-phonon coupling yields a heavy electron mass and the large universal value of $A/\gamma^2=1.0\times10^{-5}\,\mu\Omega$ cm (mol K/mJ)^2, which classify these substances as members of the heavy fermion group. Since the crystal structure of $YInCu_4$ belongs to the same category as the C15 structure, we examined the mechanism of mass enhancement. We estimated $A/\gamma^2=3.4\times10^{-4}\,\mu\Omega$ cm (mol K/mJ)^2 for $YInCu_4$ from $A=0.0042\,\mu\Omega$ cm/K^2 and $\gamma=3.5$ mJ/mol K^2. This value is much larger than that of the heavy fermion group, i.e., $A/\gamma^2=1.0\times10^{-5}$ $\mu\Omega$ cm (mol K/mJ)^2. Thus, in a certain sense, the γ is not enhanced but rather conventional. The T^2 dependence of the resistivity probably does not originate in the strong electron-phonon coupling.

In conclusion, $YInCu_4$ is presumably classified as a semimetal, and the origin of the anomalies in the resistivity and C/T may be explained within the framework of semimetallic substances. A detailed analysis of the transport properties for not only $YInCu_4$ but also the other $RInCu_4$ compounds will be published elsewhere.[8] Finally, we would like to comment that the anomalous magnetic properties observed in $YbInCu_4$, $GdInCu_4$, etc. may be closely correlated with the semimetallic behavior of the conduction band of the system.

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References

1) I. Felner and I. Nowik: Phys. Rev. B **33** (1986) 617.
2) I. Felner, I. Nowik, D. Vaknin, Ulrike Potzel, J. Moser, G. M. Kalvius, G. Wortmann, G. Schmiester, G. Hilscher, E. Gratz, C. Schmitzer, N. Pillmayr, K. G. Prasad, H. de Waard and H. Pinto: Phys. Rev. B **35** (1987) 6956.
3) H. Nakamura, K. Nakajima, Y. Kitaoka, K. Asayama, K. Yoshimura and T. Nitta: J. Phys. Soc. Jpn. **59** (1990) 28.
4) H. Nakamura, K. Ito, H. Wada and M. Shiga: Physica **B** in press.
5) K. Takegahara and T. Kasuya: J. Phys. Soc. Jpn. **59** (1990) 3299.
6) H. Nakamura, A. Uenishi and M. Shiga: unpublished.
7) H. Muller, E. Bauer, E. Gratz, K. Yoshimura, T. Nitta and M. Mekata: J. Magn. & Magn. Mater. **76 & 77** (1988) 159.
8) K. Ito, H. Nakamura and M. Shiga: *in preparation*.
9) A. Ochiai, T. Suzuki and T. Kasuya: J. Phys. Soc. Jpn. **59** (1990) 4129.
10) K. Miyake, T. Matsuura and C. M. Varma: Solid State Commun. **71** (1989) 1149.