Magnetic and transport properties in the low-carrier system $Gd_{1-x}Lu_xInCu_4$

1994 J. Phys.: Condens. Matter 6 6801

(http://iopscience.iop.org/0953-8984/6/34/012)

J. Phys.: Condens. Matter 6 (1994) 6801–6813. Printed in the UK

Magnetic and transport properties in the low-carrier system $Gd_{1-x}Lu_xInCu_4$

H Nakamura, K Ito and M Shiga
Department of Metal Science and Technology, Kyoto University, Kyoto 606-01, Japan

Received 29 March 1994, in final form 25 May 1994

Abstract. $GdInCu_4$, which forms the cubic C15b-type crystal structure, is ordered antiferromagnetically at a very low temperature of 6.9 K in spite of the large antiferromagnetic correlation expected from the large negative paramagnetic Curie temperature, −45 K. The electrical resistivity shows anomalous temperature dependence, that is, with increasing temperature, the resistivity reaches a maximum at about 80 K and decreases monotonically at higher temperatures. To study the magnetic and transport properties of $GdInCu_4$ in more detail, we measured systematically the magnetic susceptibility, electrical resistivity and thermal expansion of the $Gd_{1-x}Lu_xInCu_4$ system. The AC susceptibility and Hall coefficient of $GdInCu_4$ were also measured. The results indicate that $Gd_{1-x}Lu_xInCu_4$ is a low-carrier system and that the variation of carrier concentration caused by the change of unit cell volume and temperature affects sensitively both transport and magnetic properties. An analysis of specific heat and AC susceptibility suggests that an antiferromagnetic short-range magnetic correlation in $GdInCu_4$ persists at temperatures higher than T_N. The concept of magnetic frustration is applied to explain the anomalous magnetic properties.

1. Introduction

The intermetallic compounds, RCu_5 (R= some heavy rare-earth elements and uranium), crystallize into the cubic $AuBe_5$ (C15b)-type structure, where the magnetic atoms are located at face-centred cubic (FCC) positions. Characteristic magnetic properties have been found in *antiferromagnetic* substances; for example, a metamagnetic transition in $DyCu_5$ [1] and non-collinear spin arrangement and an unidentified phase transition in UCu_5 [2, 3]. Although the origin of these anomalous properties has not been completely understood, the degeneracy of the antiferromagnetic ground state in a high-symmetry lattice is probably responsible for the interesting properties. The system where one of the Cu atoms in RCu_5 is replaced by another element, $RTCu_4$, belongs to the same category from the magnetic viewpoint when the crystal structure belongs to the same space group. Therefore the $RTCu_4$ system is eminently suitable to investigate the magnetism in a high-symmetry crystal.

On the other hand, the $RInCu_4$ system has recently attracted considerable interest [4–20]. In particular, $YbInCu_4$ has been extensively studied in connection with the instability of the Yb valence [4–16]. Our detailed experimental study of $YInCu_4$ and $LuInCu_4$ [20] and a band calculation for $LuInCu_4$ [15] showed that the $RInCu_4$ system can be considered as a semimetal with small carrier density. Therefore, $RInCu_4$ is an appropriate system to study the magnetism in a low-carrier system. In the study of *classical* rare-earth intermetallic compounds, it is generally accepted that the long-range indirect interaction via the conduction electrons, i.e., the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, is the most relevant mechanism for the description of the magnetic properties. However, a short-range superexchange interaction could be dominant in the low-carrier system.

0953-8984/94/346801+13$19.50

In a previous paper, we studied the magnetic and transport properties of $GdInCu_4$ in order to investigate the antiferromagnetism in a high-symmetry frustrated lattice with low carrier density [18]. We selected this system since the effect of the crystalline field can be excluded because of the S state of the Gd^{3+} ion. We found that $GdInCu_4$ shows an antiferromagnetic-like transition at a very low temperature of 6.9 K in spite of the relatively large antiferromagnetic correlation expected from the paramagnetic Curie temperature, −45 K. The low-temperature specific heat shows a clear λ-type peak at $T_N = 6.9$ K. The electrical resistivity exhibits a sharp peak at T_N and, in addition, a broad hump around 80 K and negative temperature dependence above this temperature. The resistivity at the lowest temperature is fairly large.

In order to investigate the origin of the magnetic and transport properties of $GdInCu_4$, we studied the effect of the substitution of non-magnetic Lu for Gd. In this paper, we will report the results of magnetic and transport measurements of the $Gd_{1-x}Lu_xInCu_4$ system. The transport properties will be explained within the framework of a semimetal. The anomalous magnetic properties of $GdInCu_4$ will be discussed in connection with the FCC configuration of the magnetic atoms.

2. Experimental procedures

The samples were prepared in an arc furnace under an Ar atmosphere. The purities of the mother metals, Gd, Lu, In and Cu, were 99.9%, 99.9%, 99.99% and 99.999% respectively. The weight of each sample was 8–10 g. In melting, the samples were turned upside down and remelted two or three times to improve homogeneity. After the melting, each sample was wrapped in a Ta foil and annealed in an evaculated silica tube for one week. The annealing temperatures were 700–750 °C. After the annealing, x-ray powder diffraction measurements were carried out at room temperature in order to identify the crystal structure and to estimate the lattice parameter. The superlattice reflections indexed to (200) and (420), which are the evidence of the C15b structure, were observed in the x-ray powder diffraction patterns for all the samples used in the present study. No phases other than C15b phase were detected by the x-ray diffraction.

The magnetic (DC) susceptibility was measured with a magnetic torsion balance under a field of 8.28 kOe. The AC susceptibility was measured by a conventional AC bridge method with a maximum driving field of 6.6 Oe at 200 Hz. The electrical resistivity was measured using a four-probe method between 4.2 K and 1000 K. The Hall coefficient was measured in a field of 15 kOe with electric current of 70 mA. A five-probe method was used to adjust zero voltage in zero field. The thermal expansion was measured by a conventional differential-transformer-type dilatometer.

3. Experimental results

3.1. Lattice parameter and thermal expansion

The single phase of the cubic C15b crystal structure was confirmed for all $Gd_{1-x}Lu_xInCu_4$ compounds. Figure 1 shows the concentration dependence of the lattice parameter at room temperature together with the result for $YInCu_4$ [19]. The lattice parameter decreases linearly with decreasing Gd concentration, obeying approximately Vegard's law.

We measured the thermal expansion curves of $GdInCu_4$ and $LuInCu_4$ from 4.2 K to 300 K and observed the same monotonic curves for both compounds. This indicates that

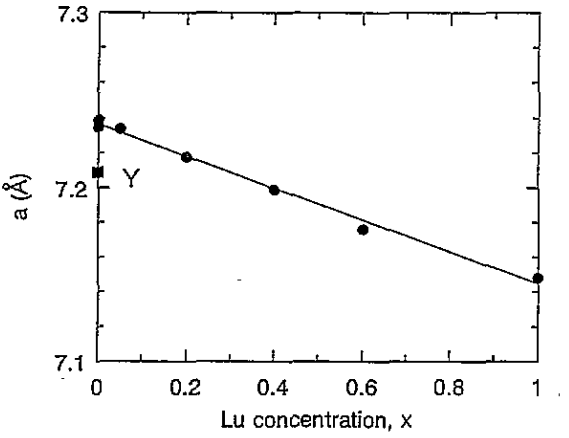

Figure 1. The lattice parameter of $Gd_{1-x}Lu_xInCu_4$ at room temperature as a function of Lu concentration. The value for $YInCu_4$ is also shown by a square. The straight line indicates Vegard's law.

the thermal expansion is dominated only by the lattice vibrations and is independent of the nature of the rare earth in the $Gd_{1-x}Lu_xInCu_4$ system. The thermal expansion coefficient estimated from the linear part above 150 K is 1.47×10^{-5} K^{-1}.

3.2. Magnetic susceptibility

Figure 2 shows the temperature dependence of the inverse magnetic susceptibility, $1/\chi$, of the $Gd_{1-x}Lu_xInCu_4$ compounds. The susceptibility follows the Curie–Weiss law down to low temperatures. The effective paramagnetic moment, $P_{\rm eff}$, and the paramagnetic Curie temperature, $\theta_{\rm p}$, are shown in figures 3(a) and (b), respectively, as a function of Lu content, x. $P_{\rm eff}$ decreases linearly with x, indicating the simple dilution of the moment. The moments per Gd atom have values within the range of $8.28 \pm 0.13\mu_{\rm B}$ for all the compounds. These are in reasonable agreement with the calculated value for Gd^{3+}, $7.94\mu_{\rm B}$. With increasing Lu content, $-\theta_{\rm p}$ also decreases, indicating that the antiferromagnetic interaction becomes weak. However, the concentration dependence is weaker than the simple dilution.

As was shown in a previous paper [18], a clear antiferromagnetic-like transition was observed at 6.9 K for $GdInCu_4$, especially in the specific heat. The low-temperature part of the susceptibility for $GdInCu_4$ is shown in figure 4. It should be noted that a peak, which is usually observed for a Heisenberg antiferromagnet, was not observed. It only deviates from the Curie–Weiss law below $T_{\rm N}$. Since the susceptibility was measured in a relatively high magnetic field of 8.28 kOe, we also measured the AC susceptibility with a maximum oscillating field of 6.6 Oe to see the response to a very low field. The result is also shown in figure 4. The temperature dependence is different from that of the DC susceptibility. Below about 20 K, the AC susceptibility deviates from the Curie–Weiss law and saturates to a constant value. This suggests that an antiferromagnetic correlation starts to develop at a much higher temperature than the long-range ordering temperature, and that the antiferromagnetic correlation is easily suppressed by the field. Furthermore, even in the very low field, a clear anomaly was not observed at $T_{\rm N}$. This suggests that the spin arrangement in the ordered state is not simple.

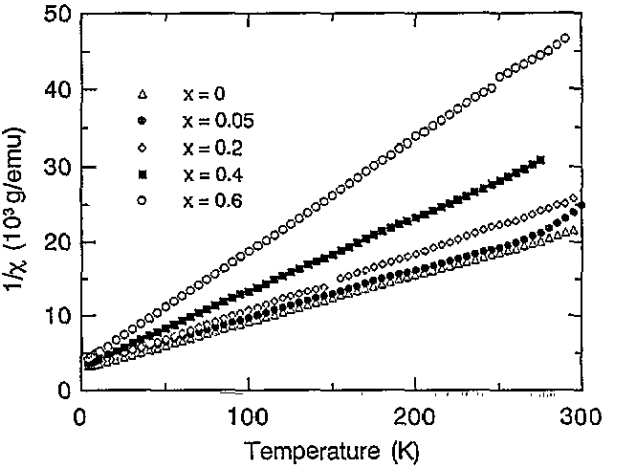

Figure 2. The temperature dependence of the inverse susceptibility of $Gd_{1-x}Lu_xInCu_x$ with $x = 0$, 0.05, 0.2, 0.4 and 0.6. Measurements were made in a field of 8.28 kOe.

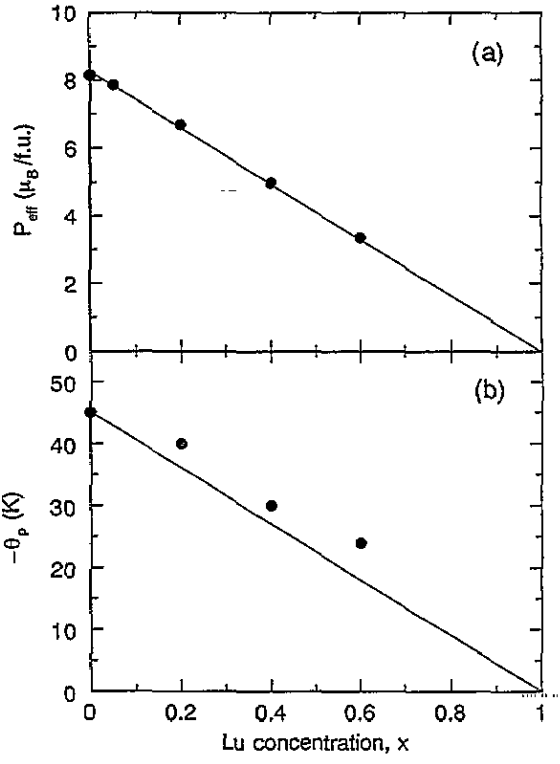

Figure 3. (a) The effective paramagnetic moment of $Gd_{1-x}Lu_xInCu_4$ as a function of Lu concentration. (b) The paramagnetic Curie temperature of $Gd_{1-x}Lu_xInCu_4$ as a function of Lu concentration. The straight lines indicate the relation expected from simple dilution.

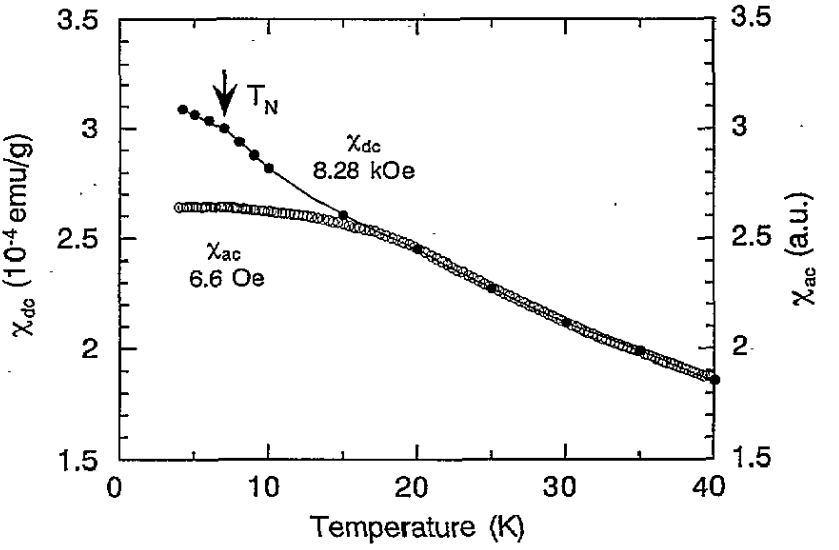

Figure 4. The low-temperature part of the DC susceptibility of GdInCu$_4$ measured in a field of 8.28 kOe and the AC susceptibility measured with a maximum oscillating field of 6.6 Oe. The arrow indicates the magnetic ordering temperature.

3.3. Electrical resistivity

The temperature dependence of the electrical resistivity of $Gd_{1-x}Lu_xInCu_4$ is presented in figure 5(a). The absolute values reduces with decreasing Gd concentration. Although a sharp peak due to the magnetic ordering was not observed for $x > 0$, the resistivity increases rapidly at very low temperatures for $x = 0.2$ as shown in figure 5(b). This indicates that the ordering temperature decreases with increasing x and becomes lower than 4.2 K for $x \geqslant 0.2$. At high temperatures, a negative dependence on temperature was observed for $x \leqslant 0.4$. The temperature of the maximum, which will be denoted as T_{max}, shifts towards higher temperatures with increasing x. For $x = 0$, 0.2 and 0.4, T_{max} is estimated to be 80, 150 and 400 K, respectively. For $x > 0.4$, the electrical resistivity increases monotonically up to the highest temperature. The resistivity, however, increases rather rapidly at low temperatures and gradually at high temperatures.

In figure 6, the residual resistivity, which is an extrapolated value to 0 K neglecting the anomaly around transition points, is plotted as a function of Lu content. The residual resistivity decreases rapidly with increasing x. This suggests that the concentration dependence of the residual resistivity is dominated not by impurity scattering but by another mechanism.

3.4. Hall coefficient

We measured the temperature dependence of the Hall coefficient for GdInCu$_4$ from 77 K to 300 K; this is shown in figure 7. The Hall coefficient is positive in the entire range of the measurement. Neglecting the effect of demagnetizing field, the Hall coefficient, R_H, for magnetic materials is usually expressed as

$$R_H = R_0 + R_s\chi \tag{1}$$

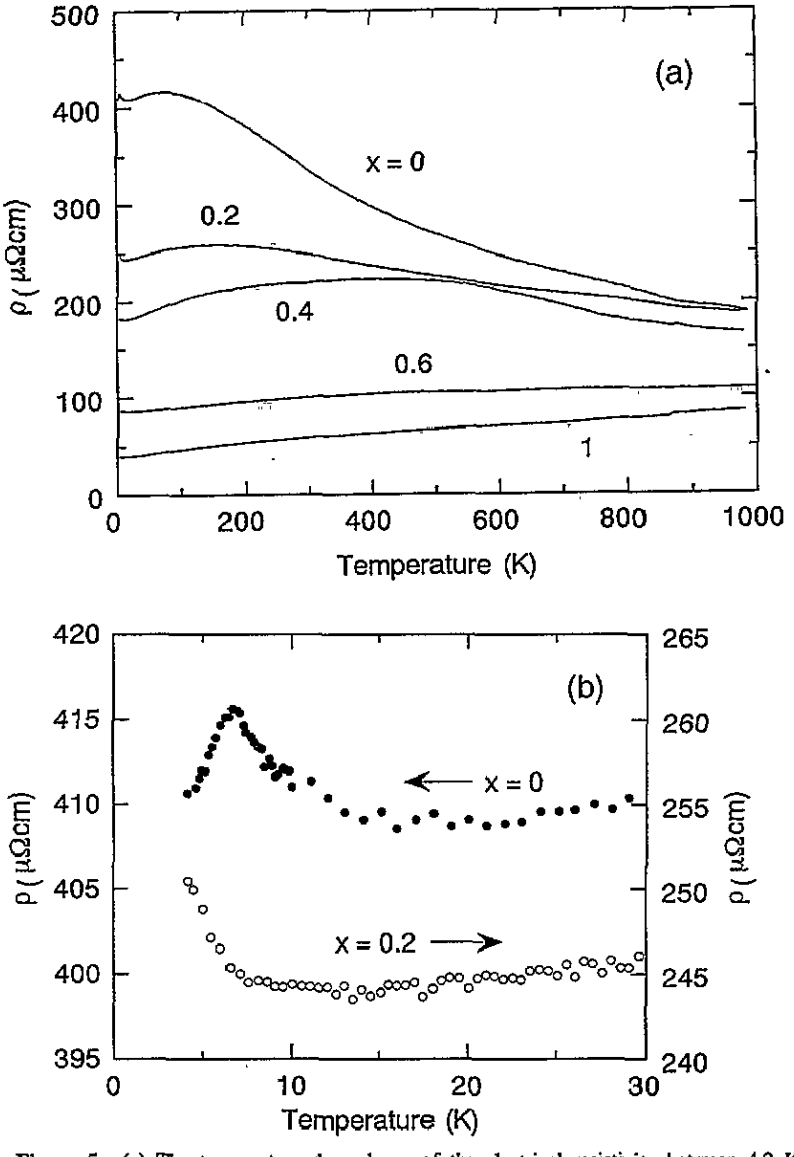

Figure 5. (a) The temperature dependence of the electrical resistivity between 4.2 K and 1000 K for $Gd_{1-x}Lu_xInCu_4$ with $x = 0$, 0.2, 0.4, 0.6 and 1. (b) The electrical resistivity at low temperatures for $GdInCu_4$ and $Gd_{0.8}Lu_{0.2}InCu_4$.

where R_0 and R_s are the normal and extraordinary Hall coefficients, respectively. For Gd compounds, a large extraordinary term is expected. For simplicity, we assume that R_0 and R_s are independent of temperature, although this assumption is not appropriate, especially for a low-carrier system. Experimental data were fitted to an expression of the form

$$R_H = R_0 + A/(T - \theta_p) \tag{2}$$

where θ_p is the paramagnetic Curie temperature deduced from the susceptibility, and A a constant. The best-fit curve with $R_0 = 0.010$ cm^3 C^{-1} and $A = -0.172$ cm^3 C^{-1} K^{-1} is

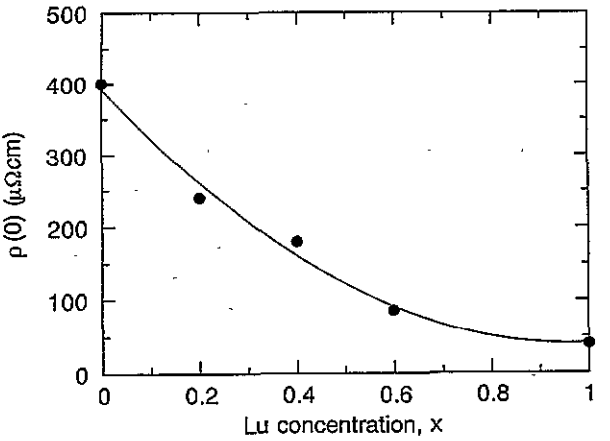

Figure 6. The concentration dependence of the residual resistivity, which is the extrapolated value of resistivity to 0 K from higher temperature neglecting the anomaly around the transition point. The solid curve is a guide for the eye.

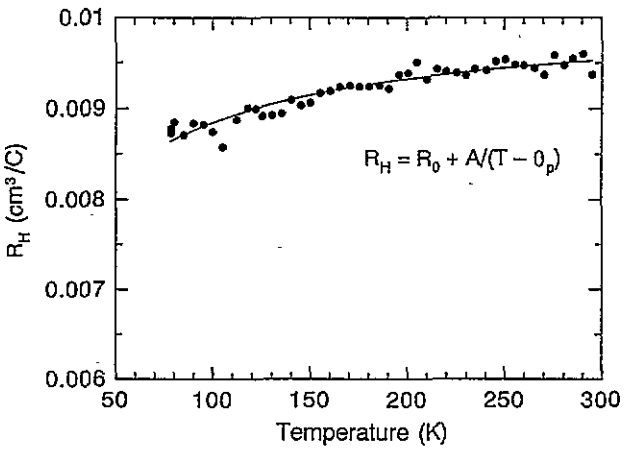

Figure 7. The temperature dependence of the Hall coefficient for GdInCu$_4$. The solid line is the best fit to an expression of the form $R_H = R_0 + A/(T - \theta_p)$, with $R_0 = 0.010$ cm^3 C^{-1}, $A = -0.172$ cm^3 C^{-1} K^{-1} and $\theta_p = -45$ K.

shown by a solid line in the figure. Assuming the single-carrier model, the carrier density n is estimated from $R_0 = 1/(ne)$ to be 0.62×10^{17} cm^{-3}, which roughly corresponds to 6×10^{-6} per formula unit. Starting from the assumption of a single type of carrier for this system, the small value of n indicates that GdInCu$_4$ is a low-carrier system.

4. Discussion

GdInCu$_4$ looks like a metallic antiferromagnet with well defined Gd local moments. We believe, however, that the small carrier concentration dominates the overall physical

properties. Therefore, we first discuss the transport properties, especially the origin of the anomalous temperature dependence of the resistivity.

4.1. Transport properties

4.1.1. Semimetallic behaviour. First, we discuss the origin of the broad peak and negative temperature dependence in the resistivity curve of $Gd_{1-x}Lu_xInCu_4$. The contribution of magnetic scattering of conduction electrons on Gd spins is ruled out as the origin of the anomalous resistivity because a similar behaviour was observed for $YInCu_4$ with no magnetic elements, for which T_{max} is ~ 270 K [19]. Therefore, the temperature dependence of the resistivity can probably be ascribed to the characteristic band structure of the $RInCu_4$ system. We try to explain the variation of the resistivity of $Gd_{1-x}Lu_xInCu_4$ using a semimetallic band picture. According to a band calculation [15], $LuInCu_4$ is classified as a semimetal with very small electron and hole Fermi surfaces. The energy bands have a small indirect overlap of the conduction bands at the X point and the valence bands at the W point. We assumed that the band overlap decreases with increasing unit cell volume and, as a result, the carrier concentration decreases. If the band overlap and Fermi energy, i.e., energy measured from the bottom or top of each band, are smaller than or comparable to k_BT at room temperature, a notable change in carrier concentration is expected on raising the temperature. On the other hand, if the band overlap and the Fermi energy are much larger than k_BT, the number of carriers is described by fully degenerate Fermi statistics, which causes metallic behaviour of resistivity. From the Hall coefficient, we estimated the carrier concentration of $GdInCu_4$ to be 0.67×10^{17} cm^{-3} based on the single-carrier model. This small value is direct evidence that $GdInCu_4$ is a low-carrier system. It should be noted, however, that the single-carrier model is not justified because the $RInCu_4$ system may be a compensated semimetal and the carrier number given above may be underestimated. It should be also noted that, although we neglect the temperature dependence of the normal Hall coefficient R_0, in other words, that of carrier concentration, the variation of carrier concentration with temperature is responsible for the negative dependence of resistivity on temperature. Anyhow, at least, the carrier concentration of $GdInCu_4$ is expected to be less than that of $YInCu_4$ because the lattice parameter of $GdInCu_4$ is larger than that of $YInCu_4$. For $YInCu_4$ and $LuInCu_4$, we estimated the number of electrons and holes to be 2.6×10^{19} cm^{-3} and 3.5×10^{20} cm^{-3} at 10 K, respectively, assuming compensation of electrons and holes [20].

4.1.2. An interpretation of the residual resistivity. Assuming that these materials are compensated semimetals, the conductivity, $\sigma(T) = 1/\rho(T)$, is given by

$$\sigma(T) = en(T)\mu(T) \tag{3}$$

where $n(T)$ is the number of electrons or holes and $\mu(T)$ the sum of electron and hole mobilities. If $\mu(T)$ is independent of the Lu concentration x, the conductivity at 0 K, $\sigma(0)$, is proportional to the carrier concentration at 0 K, $n(0)$. On the other hand, if the resistivity hump is caused by the temperature dependence of $n(T)$, k_BT_{max} is expected to be of the order of the Fermi energy, E_F, which is also given as a function of $n(0)$, because T_{max} roughly corresponds to the boundary temperature between the low-temperature range described by fully degenerate statistics and the high-temperature range described by partially degenerate statistics. In figure 8, we plot T_{max} against $\sigma(0)$, which is the inverse of the specific resistivity plotted in figure 6, together with the data of $YInCu_4$ [19]. We found an apparent correlation between T_{max} and $\sigma(0)$. This suggests that all the features of resistivity,

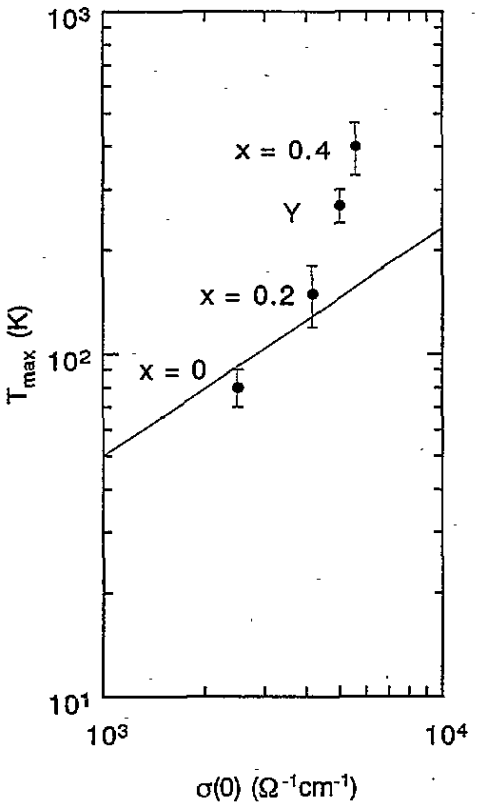

Figure 8. T_{max} against $\sigma(0)$ on a logarithmic scale for $Gd_{1-x}Lu_xInCu_4$ with $x = 0$, 0.2 and 0.4 and $YInCu_4$, where T_{max} is the temperature where the resistivity reaches a maximum and $\sigma(0)$ the conductivity at 0 K, which is the inverse of the residual resistivity. The straight line indicates the relation $T_{max} \propto n^{2/3}$.

i.e., temperature and concentration dependence, are dominated by the variation of the carrier concentration.

When one assumes parabolic bands, for which $E_F \propto n^{2/3}$, we expect $T_{max} \propto \sigma(0)^{2/3}$. In fact, T_{max} shows a stronger dependence on $\sigma(0)$, as seen in figure 8. This is not surprising because the model is too simple. In particular, the concentration dependence of $\mu(T)$ should be taken into account, because large differences in mobilites were actually observed between $YInCu_4$ and $LuInCu_4$ [20]. In the above discussion, we neglected the magnetic contribution and the electron–phonon interactions. The latter is possibly important because strong electron–phonon coupling is often observed in semimetallic systems and was actually suggested for $YInCu_4$ [19,20].

Table 1. The magnetic ordering and paramagnetic Curie temperatures of antiferromagnetic Gd compounds with the C15b-type crystal structure.

	T_N (K)	θ_p (K)	$-\theta_p/T_N$	Reference
$GdAgCu_4$	9	−7	0.78	[21]
$GdAuCu_4$	10	−13	1.3	[22]
$GdInCu_4$	6.9	−45	6.5	—

4.2. Magnetic properties

4.2.1. The exchange mechanism. The magnetic properties of $GdInCu_4$ are characterized by the large ratio of $-\theta_p/T_N$. In table 1, we tabulate θ_p and T_N of *antiferromagnetic* $GdTCu_4$ compounds with the same crystal structure [21, 22]. The ratio, $-\theta_p/T_N$, is nearly unity for all compounds except $GdInCu_4$. The ratio comes to around one to two for most of the metallic Gd compounds, for which the magnetism is described by the indirect RKKY interaction between localized spins via conduction electrons. Actually, the magnetism of $GdAgCu_4$ and $GdAuCu_4$ was explained well in terms of the RKKY theory [21, 22]. On the other hand, $-\theta_p/T_N$ is very large for $GdInCu_4$. In the localized moment system, θ_p represents the magnitude of the exchange interaction between spins. Therefore, an antiferromagnetic interaction is expected to be comparatively large in $GdInCu_4$. It is difficult to understand such a behaviour in terms of the RKKY model because the present system is semimetallic with a very small concentration of conduction electrons. Naively, with the carrier concentration approaching zero, the RKKY interaction becomes ferromagnetic-like because the period of the RKKY oscilation becomes long, and the absolute value becomes small. Therefore, the RKKY interaction cannot explain the much larger antiferromagnetic interaction of $GdTCu_4$ than of the other $GdTCu_4$ compounds.

Let us consider other mechanisms of interaction in the present system. The superexchange interaction via p–f and d–f mixing may be a candidate. Most simply, a direct mixing between Gd 4f and In 5p or Cu 3p, 3d states is considered. However, this may be too small because the 4f wave functions of Gd are shielded by the 6s and 5d wave functions and then mixing between 4f and the other states is small. It should be noted that, according to the band calculation of $LuInCu_4$ [15], the conduction bands above E_F are derived mainly from the Lu 5d state. Therefore, we consider, as higher-order interactions, a combined mechanism of electron (charge) transfer from the Gd 5d state to, for example, the In 5p state and Gd intra-atomic exchange interaction between 5d and 4f states. In this case, degenerate In 5p wave functions in tetrahedral interstices bond nearest Gd atoms. A similar mechanism has been proposed as the origin of a dominant exchange interaction for Eu chalcogenides [23]. Thus, we speculate that some *short-range* mechanism produces a large antiferromagnetic interaction in $GdInCu_4$, being different from other $GdTCu_4$ compounds. For a further understanding, theoretical considerations based on the actual band structure would be useful.

4.2.2. Analysis of specific heat. In order to obtain information on the origin of the large value of $-\theta_p/T_N$, an analysis of specific-heat data [18] is presented. The magnetic entropy, S_m, of $GdInCu_4$ is shown in figure 9. The entropy at a given temperature was calculated by integrating C_m/T from 0 K to the temperature, where the magnetic contribution of specific heat, C_m, was estimated by subtracting the specific heat of $YInCu_4$ [19] from that of $GdInCu_4$. We assumed a linear temperature dependence of C_m/T between 0 K and 1.5 K. Another analysis using the data of $LuInCu_4$ [24] does not lead to a substantial difference. The entropy, S_m, reaches 11 J mol^{-1} K^{-1} at T_N, increases gradually above T_N and does not saturate even at 20 K ($\sim 3T_N$). For a Heisenberg system, the magnetic entropy should attain its full value $R\ln(2J+1)$ at high temperature, where R is the molar gas constant. For Gd compounds, because of their S state with zero orbital moment, the ground-state multiplet, $J = \frac{7}{2}$, is not influenced by the crystalline electric field. The full entropy, $R\ln 8 = 17.3$ J mol^{-1} K^{-1}, therefore, is attained in principle just above the ordering temperature. Usually, the magnetic entropy above T_N attains at least 80–90% of $R\ln 8$ for conventional Gd compounds [25]. It should be noted that the entropy of $GdInCu_4$ is anomalously small, reaching 60% just above T_N and attaining only 75% at $3T_N \simeq 20$ K. This

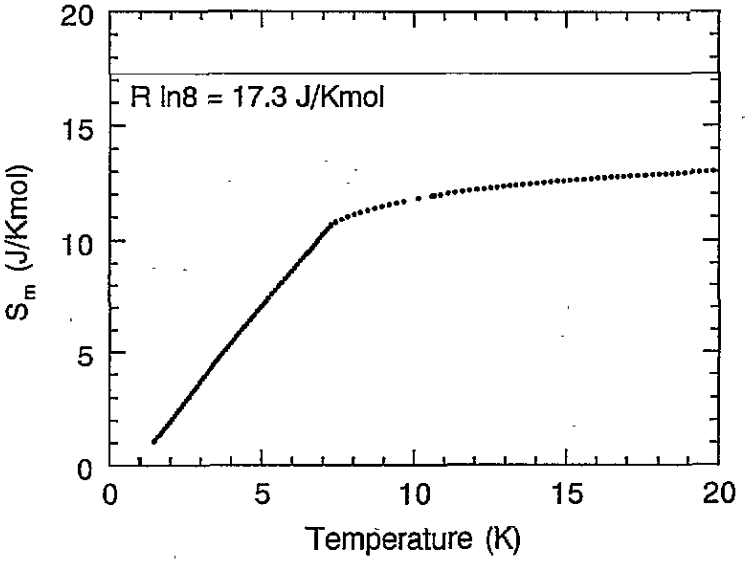

Figure 9. The temperature dependence of the magnetic entropy of GdInCu$_4$ estimated from the specific heat of GdInCu$_4$ and YInCu$_4$. The method of analysis is described in the text. The straight line indicates the expected value for a Heisenberg magnet with $J = \frac{7}{2}$.

indicates that short-range magnetic correlations persist even at temperatures much higher than T_N. This idea reasonably explains the anomalous behaviour of the AC susceptibility measured in low fields, which starts to deviate from Curie–Weiss behaviour at a temperature much higher than T_N.

4.2.3. An interpretation in terms of the frustration in the FCC lattice. In a first approximation, the value of $-\theta_p/T_N$ can be explained in terms of the Heisenberg model within a molecular-field approximation. The ratio can be formally reproduced for the FCC lattice by using two parameters of effective nearest- and next-nearest-neighbour exchange integrals, J_1 and J_2, if the antiferromagnetic modulation vector, $\boldsymbol{Q}$, in the ordered state is known [26]. However, the molecular-field approximation gives no information on the characteristic behaviour above T_N such as the reduced value of the entropy and the anomalous susceptibility.

Hence, we introduce another concept, the frustration among nearest-neighbour antiferromagnetic interactions, as a possible explanation of the large $-\theta_p/T_N$ of GdInCu$_4$. Usually, the effect of frustration on the phase transition appears as a depression of the transition temperature or successive phase transitions. Ramirez [27] discussed the magnitude of $-\theta_p/T_N$ for magnetic insulators in terms of the frustration effect. The ratio, $-\theta_p/T_N$, exceeds 10 in some insulators that have characteristic crystal structures such as the triangular lattice. For example, a garnet $Gd_3Ga_5O_{12}$ has a huge $-\theta_p/T_N > 100$ [27, 28]. For insulators containing Gd, however, $|\theta_p|$ is usually small ($\theta_p = -2.3$ K for $Gd_3Ga_5O_{12}$) because of the lack of the RKKY mechanism whereas in metallic compounds, $|\theta_p|$ can be large and $-\theta_p/T_N$ rarely exceeds five. The largest value ever known is 5.3 for $GdAl_3$ ($\theta_p = -90$ K, $T_N = 17$ K) [29] and Gd_2Zn_{17} ($\theta_p = -53$ K, $T_N = 10$ K) [30]. Although it is reasonably accepted that the long-range RKKY interaction is dominant in these compounds, the effects of frustration were also suggested. Coles *et al* [31] pointed out that the frustration of $GdAl_3$ depresses the onset of antiferromagnetism, because, in $GdAl_3$, Gd atoms are arranged just

like a triangular lattice on the basal plane. They claimed that $GdAl_3$ possesses competing interactions, which suppress long-range ordering from an expected onset temperature of about 100 K to the observed T_N of 17 K, and that as a result short-range order effects occur below 100 K; these were revealed by an electron spin resonance (ESR) measurement. In $GdInCu_4$, a larger value of $-\theta_p/T_N$ than $GdAl_3$ was obtained.

We assume for $GdInCu_4$ that (i) the RKKY and the classical dipolar interactions are negligibly small and (ii) the superexchange mechanism between Gd atoms via In is dominant. In the compound, Gd and In form a zinc-blende-type superlattice. It should be noted that an In atom is located at the centre of a tetrahedron formed by four Gd atoms, which are the nearest magnetic atoms to each other. If only the superexchange mechanism is considered, strong exchange interaction exists between nearest Gd atoms and there is no substantial coupling between next-nearest Gd atoms. Therefore, within this assumption, $GdInCu_4$ is a unique FCC system where only the nearest-neighbour antiferromagnetic interaction is present. It is well known that an antiferromagnetic nearest-neighbour interaction frustrates in the FCC lattice. Thus, we reasonably interpret the anomalous properties of $GdInCu_4$ in terms of the frustration. We speculate that, in $GdInCu_4$, the frustration depresses the static magnetic ordering down to a very low temperature from an expected onset temperature of about 45 K, and, as a result, the short-range dynamic correlation persists up to high temperatures as was suggested by the lack of magnetic entropy above T_N and the deviation from the Curie–Weiss behaviour of the susceptibility from a temperature much higher than T_N. Magnetic ordering is possibly realized by other additional long-range interactions such as the RKKY and dipolar interactions. The possible ground state in the frustrated system is a magnetically ordered state with a non-collinear structure, a spin glass, a spin-liquid state, etc. Although we do not have any information on the magnetic structure of $GdInCu_4$, the absence of the peak in the susceptibility rules out a simple antiferromagnet, and suggests a complex structure with ferromagnetic-like components. Finally, we comment that, in this system, the small carrier concentration and unique atomic configuration are necessary conditions to give rise to the frustration effects.

5. Concluding remarks

Magnetic and transport properties of the pseudoternary system $Gd_{1-x}Lu_xInCu_4$, which forms the cubic C15b crystal structure with an FCC network of rare-earth atoms, were studied. We found that this system is a low-carrier system and the transport properties are characterized by the variation of the very small carrier concentration. The anomalous transport properties were explained by assuming a semimetallic band structure, which has small electron and hole bands. Based on the fact that $GdInCu_4$ is a low-carrier system, we speculated the magnetic interaction between Gd spins, and applied the concept of frustration to explain the anomalous magnetic properties of $GdInCu_4$. The frustration depresses the static magnetic ordering down to a very low temperature from the expected ordering temperature of the order of $-\theta_p$ and, as a result, the short-range dynamic correlations persist up to high temperatures. Experiments in $GdInCu_4$ to detect the spin dynamics above T_N and to determine the spin structure below T_N would be interesting. Theoretical works based on the actual band structure are also desired.

Acknowledgments

The authors would like to acknowledge Dr H Wada for collaboration with specific heat

measurements, Mr R Iehara for much technical assistance and Mr N Shimamura and Mr K Hashi for assistance with experiments.

References

[1] Buschow K H J, van Diepen A M and de Wijn H W 1970 *J. Appl. Phys.* **41** 4609
[2] Nakamura H, Kitaoka Y, Inoue M, Asayama K and Ōnuki Y 1990 *J. Magn. Magn. Mater.* **90&91** 459
[3] Ott H R, Rudigier H, Felder E, Fisk Z and Batlogg B 1985 *Phys. Rev. Lett.* **55** 1595
[4] Felner I and Nowik I 1986 *Phys. Rev.* B **33** 617
[5] Sampathkumaran E V, Nambudripad N, Dhar S K, Vijayaraghavan R and Kuentzler R 1987 *Phys. Rev.* B **35** 2035
[6] Felner I, Nowik I,Vaknin D, Ulrike Potzel, Moser J, Kalvius G M, Wortmann G, Schmiester G, Hilscher G, Gratz E, Schmitzer C, Pillmayr N, Prasad K G, de Waard H and Pinto H 1987 *Phys. Rev.* B **35** 6956
[7] Yoshimura K, Nitta T, Mekata M, Shimizu T, Sakakibara T, Goto T and Kido G 1988 *Phys. Rev. Lett.* **60** 851
[8] Shimizu T, Yoshimura K, Nitta T, Sakakibara T, Goto T and Mekata M 1988 *J. Phys. Soc. Japan* **57** 405
[9] Nowik I, Felner I, Voiron J, Beille J, Najib A, du Tremolet de Lacheisserie E and Gratz E 1988 *Phys. Rev.* B **37** 5633
[10] Ogawa S, Suga S, Taniguchi M, Fujisawa M, Fujimori A, Shimizu T, Yasuoka H and Yoshimura K 1988 *Solid State Commun.* **67** 1093
[11] Kojima K, Hayashi H, Minami A, Kasamatsu Y and Hihara T 1989 *J. Magn. Magn. Mater.* **81** 267
[12] Nakamura H, Nakajima K, Kitaoka Y, Asayama K, Yoshimura K and Nitta T 1990 *J. Phys. Soc. Japan* **59** 28
[13] Kojima K, Nakai Y, Suzuki T, Asano H, Izumi F, Fujita T and Hihara T 1990 *J. Phys. Soc. Japan* **59** 792
[14] Itoh Y, Kadomatsu H, Sakurai J and Fujiwara H 1990 *Phys. Status Solidi* a **118** 513
[15] Takegahara K and Kasuya T 1990 *J. Phys. Soc. Japan* **59** 3299
[16] Severing A, Gratz E, Rainford B D and Yoshimura K 1990 *Physica* B **163** 409
[17] Abe S, Atsumi Y, Kaneko T and Yoshida H 1992 *J. Magn. Magn. Mater.* **104–107** 1397
[18] Nakamura H, Ito K, Wada H and Shiga M 1993 *Physica* B **186–188** 633
[19] Nakamura H, Ito K, Uenishi A, Wada H and Shiga M 1993 *J. Phys. Soc. Japan* **62** 1446
[20] Nakamura H, Ito K and Shiga M, in preparation.
[21] Takeshita T, Malik S K, El-Atttar A A and Wallace W E 1976 *AIP Conf. Proc.* **34** 230
[22] Kaneko T, Arai S, Abe S and Kamigaki K 1986 *J. Phys. Soc. Japan* **55** 4441
[23] Kasuya T 1970 *IBM J. Res. Dev.* **14** 214
[24] Pillmayr N, Bauer E and Yoshimura K 1992 *J. Magn. Magn. Mater.* **104–107** 639
[25] See, for example, Bouuvier M, Lethuillier P and Schmitt D 1991 *Phys. Rev.* B **43** 13 137
[26] See, for example, Smart J S 1963 *Magnetism* vol III, ed G T Rado and H Suhl (New York: Academic) p 63
[27] Ramirez A P 1991 *J. Appl. Phys.* **70** 5952
[28] Ramirez A P and Kleiman R N 1991 *J. Appl. Phys.* **69** 5252
[29] Buschow K H J and Fast J F 1966 *Z. Phys. Chem. Neue Folge* **50** 1
[30] Stewart A M and Coles B R 1974 *J. Phys. F: Met. Phys.* **4** 458
[31] Coles B R, Oseroff S and Fisk Z 1987 *J. Phys. F: Met. Phys.* **17** L169