Magnetic Resistivity and Kondo Effect in $Ce(Al_{1-x}Ga_x)_2$ Compounds

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ABSTRACT

Measurements of the electrical resistivity and magnetization as a function of the temperature, on polycrystalline $Ce(Al_{1-x}Ga_x)_2$ samples for $x \leq 0.4$, are presented. Evidence for the presence of the Kondo effect was deduced in the cubic phase range for $x \leq 0.15$.

Key-words: Intermetallic compounds; Kondo effect; Magnetic resistivity; Crystal field.

I - INTRODUCTION

The intermetallic compound $CeAl_2$ has been studied extensively in recent years from the experimental and theoretical point of view ([1-5] and references therein). Results indicated that the Ce moment is antiferromagneticaly modulated according to a sine wave propagation along $[1\bar{1}0]$ direction. Moreover, $CeAl_2$ presents Kondo behavior with Kondo temperature T_K comparable to the ordering one, leading to a new non magnetic ground state of the Ce^{+3} Kramers' ion. On the other hand, studies performed in hexagonal $CeGa_2$ and $Ce(Al_{0.1}Ga_{0.9})_2$ compounds [6-11] indicate that these compounds order magneticaly at the same Néel temperature T_N = 9.5K, with an incomensurate modulated structure and present traces of Kondo behavior but without the presence of minimum in the resistivity versus temperature curve above T_N. Also, in $CeGa_2$, a reduction of about 40% of the Ce moment was inferred from neutron studies (which is characteristic of the Kondo system). Similar characteristics were found in hexagonal CeAlGa [12] with T_N = 6K.In contrast to $CeAl_2$ which crystalizes in cubic $MgCu_2$ type structure (Fd3m), the Ga-rich compounds have the hexagonal AlB_2 structure (P6/mmm space group). Therefore, we use Ga for substitution at the Al site in $CeAl_2$, in order to know the evolution of the Kondo behavior and of the magnetic properties present in $CeAl_2$ with the Ga concentration. Similarly to Al, Ga is a sp^1 type, non-magnetic metal.

In this paper we present results of electrical resistivity measurements in $Ce(Al_{1-x}Ga_x)_2$ compounds. Also magnetization experiments for samples characterizations was performed.

II - EXPERIMENTAL

Polycrystalline $Ce(Al_{1-x}Ga_x)_2$ (x=0; 0.05; 0.1; 0.15; 0.2; 0.3; 0.4) and $LaAl_2$ samples were prepared by arc melting in a water cooled copper crucible under a purified argon protective atmosphere and subsequently annealed for one week at 800°C in a vacuum of 10^{-6} Torr. The phase purity was checked by X-Ray Debye-Scherrer diffractogram. Whereas in $x > 0.30$ (hexagonal) and $x < 0.10$ (cubic) no foreign phases were detectable, in $x = 0.20$ and $x = 0.15$ samples which we prepared, traces of a second phase were observed and different annealing procedures to remove them was unsuccessful. The electrical resistivity measurements were performed with the four probe DC method, between 1.8 and 300K. Measurements were performed on parallelepiped samples ($2x2x10mm^3$) cut from the bottons. In some concentrations, because of the brittleness of the samples, good mechanical quality has not been obtained and a large number of cracks were present. Cracks cause an overestimation of the sample cross-section and consequently in the electrical resistivity data and can modified the shape of the ρ versus T curves. So, results of the ρ versus T curves for $x > 0.20$ must be analysed carefully. Bulk

magnetization measurements were also carried out by using a vibrating sample magnetometer in the temperature range of 1.8 - 300K in magnetic fields H up to 13kOe. Some isotherms were obtained in an extraction magnetometer in magnetic applied fields up to 60kOe .

III - EXPERIMENTAL RESULTS

III - 1 - X-Ray

Tab.I shows the lattice parameters of $Ce(Al_{1-x}Ga_x)_2$ at room temperatures in the hexagonal and cubic phases, obtained by employing the usual Nelson-Riley extrapolation procedure. Analysis of the lattice parameters is in agreement with a trivalent state for Ce ion in the whole concentration range. We can note that the ***c/a*** values in the hexagonal phase decrease with the Al content but belong to the "intermediate ***c/a***" AlB_2 structure type [13]. On the other hand, in the cubic phase, the lattice parameters present an initial decrease with the Ga addition and tends to saturate in the mixed phase range. This decrease probably is due to the higher covalent character of the Ga atoms as compared to Al that cause Ga-Ga interactions stronger than the Al-Al one.

III - 2 - Magnetization Measurements

In Fig.1 we show the thermal variation of the magnetization for some $Ce(Al_{1-x}Ga_x)_2$ compounds performed in 1kOe applied field. No thermal hysteresis was detected for the field cooling and zero field cooling measurements. As in $CeAl_2$, the curves pass through a maximum at T_N which is the Néel's antiferromagnetic ordering temperature . The temperature of this maximum decreases in the cubic phase and increases in the hexagonal phase (see Tab. I). The magnetization M as a function of the applied magnetic field H_{ap} performed in the low temperatures regime ($T<T_N$) exhibits an inflection point characteritic of a metamagnetic type behaviour in the cubic range and, in the whole concentration range, no hysteresis was detected and is far from saturation till 60kOe. As in CeAlGa [12], the thermal variation of the reciprocal susceptibility using the Arrot plots shows that above 100K, Curie-Weiss law is observed with the slopes leading to an effective magnetic moment very close to that of the Ce^{+3} free ion for all concentrations.

III - 3 - Resistivity Measurements

Figs. 2 and 3 show the electrical resistivity ρ versus temperature curves of the $Ce(Al_{1-x}Ga_x)_2$ in the cubic and hexagonal phases respectively. As mentioned in Part-II, the resistivity in some concentration range

presents huge values probably due to the cracks in the samples; the residual resistivity ρ_0 reaches 200$\mu\Omega$cm for x = 0.40 and, at 300K, the resistivity is 900$\mu\Omega$cm. We can observe that the main characteristics of the boundary compound $CeAl_2$ are preserved in the whole cubic concentration range, i.e. a maximum at low temperature, followed by a minimum. However, this behavior vanishes with increasing Ga content and for x > 0.20, no well defined minimum in the resistivity curves is observed. So in the hexagonal concentration range, the ρ curves do not show a minimum at low temperatures, but a clear anomaly remains in this temperature range. More details of this behavior can be observed in the expanded plots of the low temperature portion of ρ versus T curves (Fig.4). In the cubic phase range an abrupt decrease of the resistivity at the low temperatures can be observed and probably is due to the development of a coherent ground state.

IV - ANALYSIS AND DISCUSSION OF THE RESULTS

As the experimental results are strongly influenced by the crystalline electric field, the analysis and discussion of the results will be subdivided in two parts: at first the cubic range (x up to 0.15) will be discussed, followed by the hexagonal range (x < 0.20). Two phases region approximately around x= 0.20, is observed.

Results of magnetization , in cubic phase range, show that the addition of Ga to $CeAl_2$ leads to a slight decrease in the Néel temperature,

indicating a decrease in the strength of the negative long range interaction coefficient J of the Ce ions. We notice however that the general features of the electrical resistivity are very similar to that of the boundary compound $CeAl_2$ that is known as a Kondo lattice system. As described by Cornut and Coqblin [3], the magnetic resistivity ρ_m as a function of the temperature, due to the scattering of the conduction electrons in the presence of the Kondo effect, is characterized by two $-J^3 \ln T$ ranges which are separated by a maximum at $T=\Delta$ (Δ is the temperature of the overall crystal field splitting).

Fig. 5 shows the magnetic contribution to the electrical resistivity, ρ_m, obtained by assuming that the phonon contribution to ρ from both $LaAl_2$ and $Ce(Al_{1-x}Ga_x)_2$ in the cubic phase range is the same. The logarithmic variation of the magnetic resistivity as a function of the temperature can be observed up to $x = 0.15$ over an appreciable temperature range. In agreement with the magnetic results, the coefficient J^3 of the lnT decreases with the Ga addition. These results indicate that the combined Kondo effect and crystal field persists with the addition of the Ga at least until $x = 0.15$. As $T_K = 1/N(E_F)EXP[-1/|J|N(E_F)]$ (where $N(E_F)$ is the density of states at the Fermi level, and J the inter-site Ruderman-Kittel-Kasuya-Yosida RKKY- interaction), the reduction of J leads to a decrease in the Kondo temperature T_K. These facts suggest that the strength of the Kondo effect progressively decreases with the addition of the Ga contents. In agreement with this assumption , we can observe (Fig.1) that the absolute value of the magnetization increases gradually with the Ga contents. This result may be inferred from a progressive screening reduction of the Ce magnetic moment.

In the hexagonal phase range (x = 0.30 and 0.40), the magnetic resistivity cannot be estimated by subtracting the ρ contribution of a non magnetic reference compound $La(Al,Ga)_2$, as usually adopted, because the absolute value of ρ cannot be calculated accurately due to the existence of microstrains in the samples. However, even without subtraction of the phonon background, we can observe that the electrical resistivity as a function of the temperature in the hexagonal phase has the same features of that in the cubic phase range. This similarity in the curves shape is reinforced when we compare the slopes of dρ/dT.

At this point it should be mentioned that the pronounced behavior of the magnetic resistivity (lnT) appears only for the case where the Kondo temperature T_K is much smaller than the crystal field splitting temperature. Hanzawa et al [14] have demonstrated that the Kondo temperature of the highest level, T_K^H in the presence of the crystal field splitting can be calculated by $T_K^H=(T_K\Delta_1\Delta_2)^{1/3}$ where, T_K is the Kondo temperature of the crystal field ground state and $\Delta_1\Delta_2$ the splitting energies of the first and second excited level, respectively. In the cubic cerium compounds, the crystal field effect gives a Γ^7 doublet and Γ^8 quartet. For $CeAl_2$, the crystal field splitting determined from the inelastic neutron measurements [15] gives Δ_1 = 100K, Δ_2 = 180K and the Kondo temperature, determined by quasi elastic line width of neutron scattering at the low temperature limit [16] is T_K = 6K. Then the calculated T_K^H = 48K which is much smaller than the crystal field splitting temperature.

On the other hand in the hexagonal Ce compound, the $J = 5/2$ multiplet is split by the crystal field into three doublets $\left|\pm\frac{1}{2}\right\rangle, \left|\pm\frac{3}{2}\right\rangle$ and $\left|\pm\frac{5}{2}\right\rangle$. Studies performed in the hexagonal compound $Ce(Al_{1-x}Ga_x)_2$ at x = 0.5 [12] indicate that the crystal field splitting determined from the neutron measurements gives Δ_1 =33K and Δ_2=212K and Θ_p = -52K. Without T_K , in spite of some incertainties, we can estimat T_K^H by using the expression $T_K^H = m\left|\Theta_P^H\right|$, where m is of the order of one. In this case , for x = 0.5 , T_K^H = 52K << Δ_2. Therefore, a higher temperature (lnT) dependence could be expected to occur for T > Δ in this concentration range. However, the magnetic resistivity ρ_m as a function of the temperature for x = 0.50 obtained by M. A. Fremy et al [12], exhibits two shoulders corresponding to the thermal population of the first and second excited CEF states. To subtract the phonon contribution in this compound, the electrical resistivity ρ was renormalized by a factor 0.45 in order to obtain, between 200 and 300K, the same slope for both LaAlGa and CeAlGa. Consequently, the above described procedure will reflect a temperature independent ρ_m in the high temperature range. For lower Ga concentrations (x=0.40 and 0.30), as the T_K^H decreases and the crystal field splitting probably varies slightly, we expecte that the magnetic resistivity ρ_m be proportional to (-lnT).

In the low temperature range, as the system presents evidence of a low Kondo system (T_K<T_N), it is quite possible that the development of ρ_{min} followed by a logarithmic dependence will be obliterated. The anomaly observed in the electrical resistivity curve near 30K can be

attributed to the resonant scattering of the hexagonal crystal field. The results of our present experimental study leads to a conclusion that, in the cubic phase range, the properties of the boundary compound $CeAl_2$ are preserved but a gradual decrease of the Kondo effect was observed. Although in the hexagonal phase range, a logarithmic variation of the magnetic resistivity is expected, as was theoretically predicted by Cornut and Coqblin [3] in the high temperature range, no clear evidence of the Kondo effect was deduced from these studies.

FIGURE CAPTIONS

Figure 1 : The thermal dependence of the magnetization of some $Ce(Al_{1-x}Ga_x)_2$ compounds. The solid lines are guide to the eyes.

Figure 2 : The thermal dependence of the electrical resistivity of $Ce(Al_{1-x}Ga_x)_2$ - cubic range.

Figure 3 : The thermal dependence of the electrical resistivity of $Ce(Al_{1-x}Ga_x)_2$ - hexagonal range.

Figure 4 : The themal dependence of ρ-expanded plot of the low temperature portion of the $Ce(Al_{1-x}Ga_x)_2$ - cubic range. Insert, the thermal dependence of ρ-expanded plot of the low-temperature portion of the $Ce(Al_{1-x}Ga_x)_2$ - hexagonal range.

Figure 5 : The magnetic contribution to the electrical resistivity ρ_m as a function of the LnT for $Ce(Al_{1-x}Ga_x)_2$.

TABLES

Table 1 : The lattice parameters and T_N for $Ce(Al_{1-x}Ga_x)_2$.

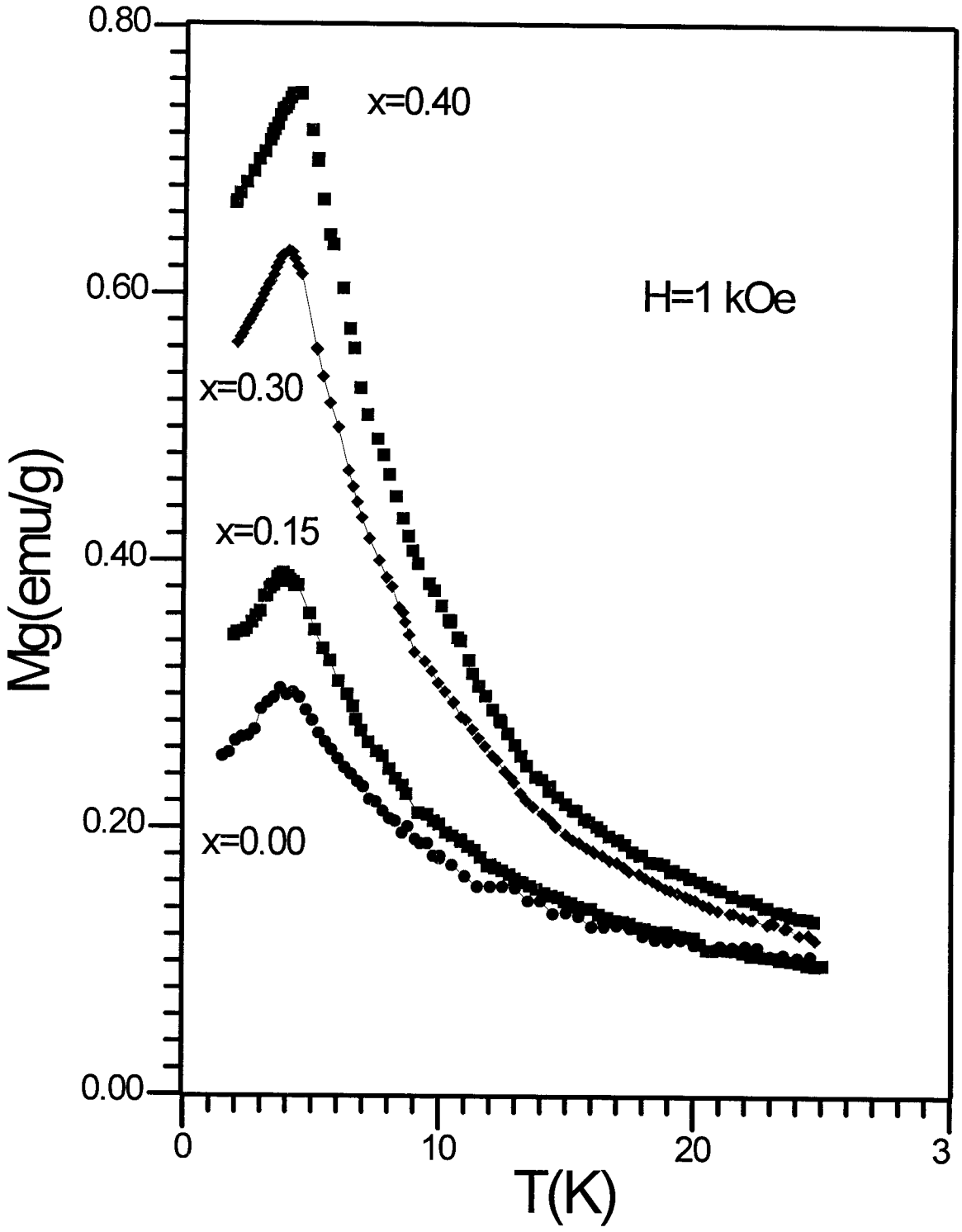
0.80
0.60
0.40
0.20
0.00
Mg(emu/g)
x=0.40
x=0.30
x=0.15
x=0.00
H=1 kOe
0
10
20
3
T(K)

Figure 1

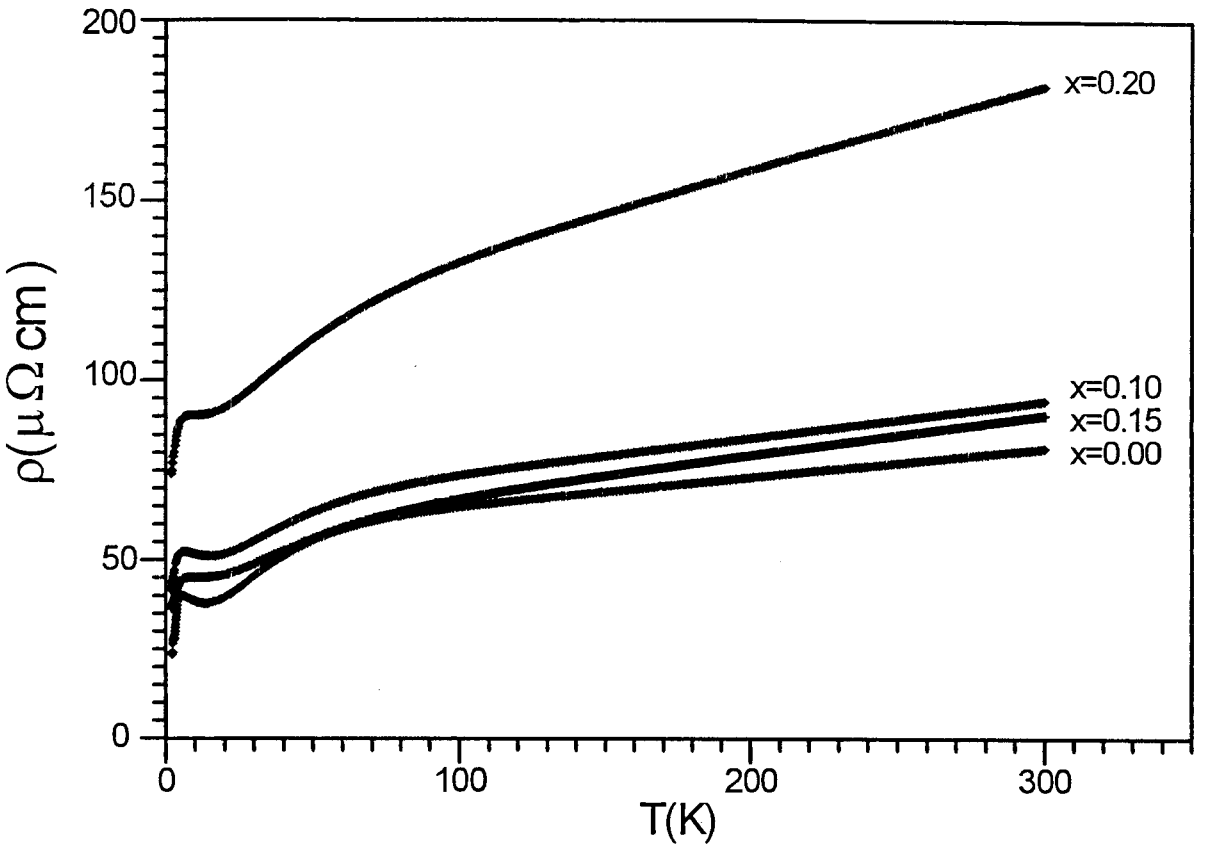

Figure 2

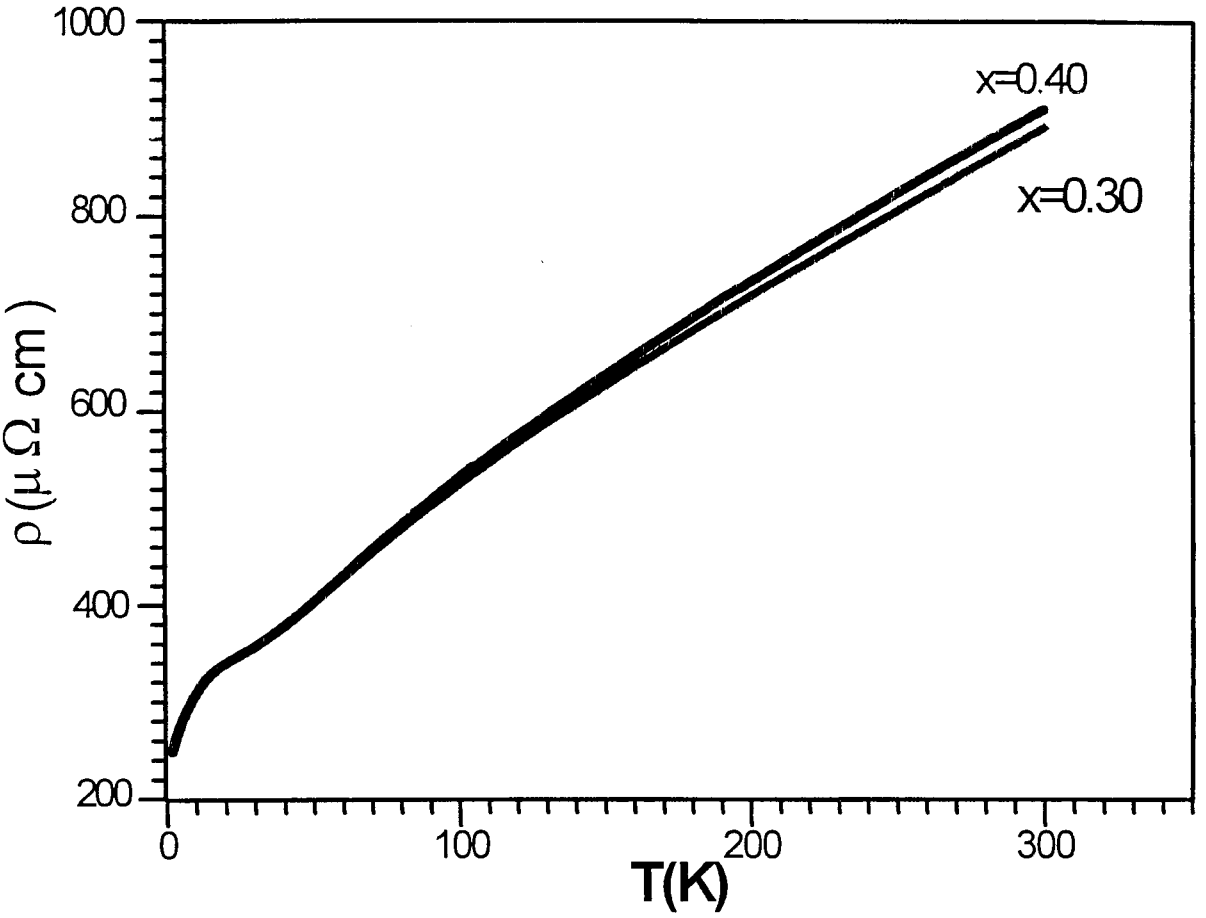

Figure 3

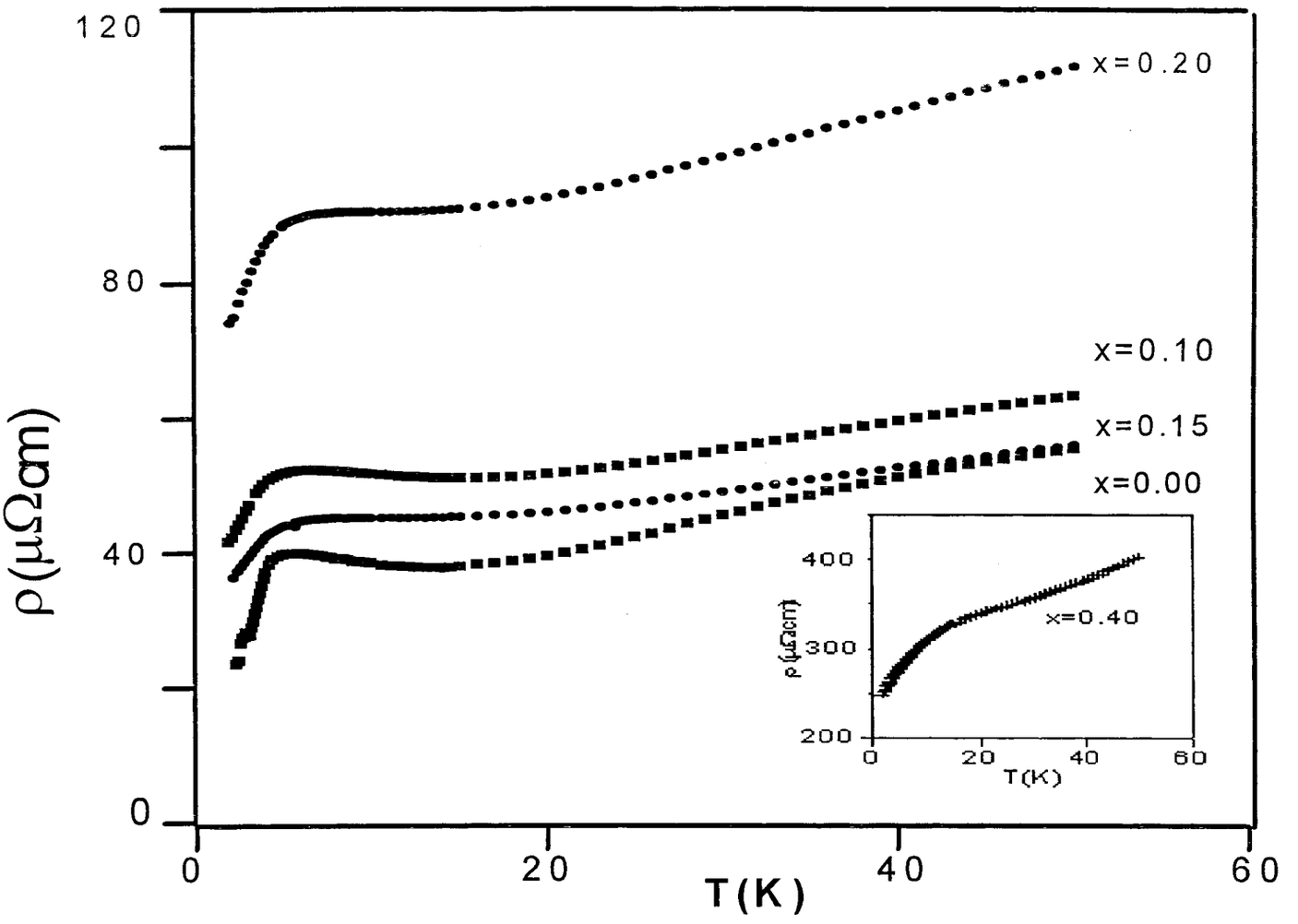
120
80
40
0
ρ(μΩcm)
0
20
40
60
T(K)
x=0.20
x=0.10
x=0.15
x=0.00
400
300
200
ρ(μΩcm)
x=0.40
0
20
40
60
T(K)

Figure 4

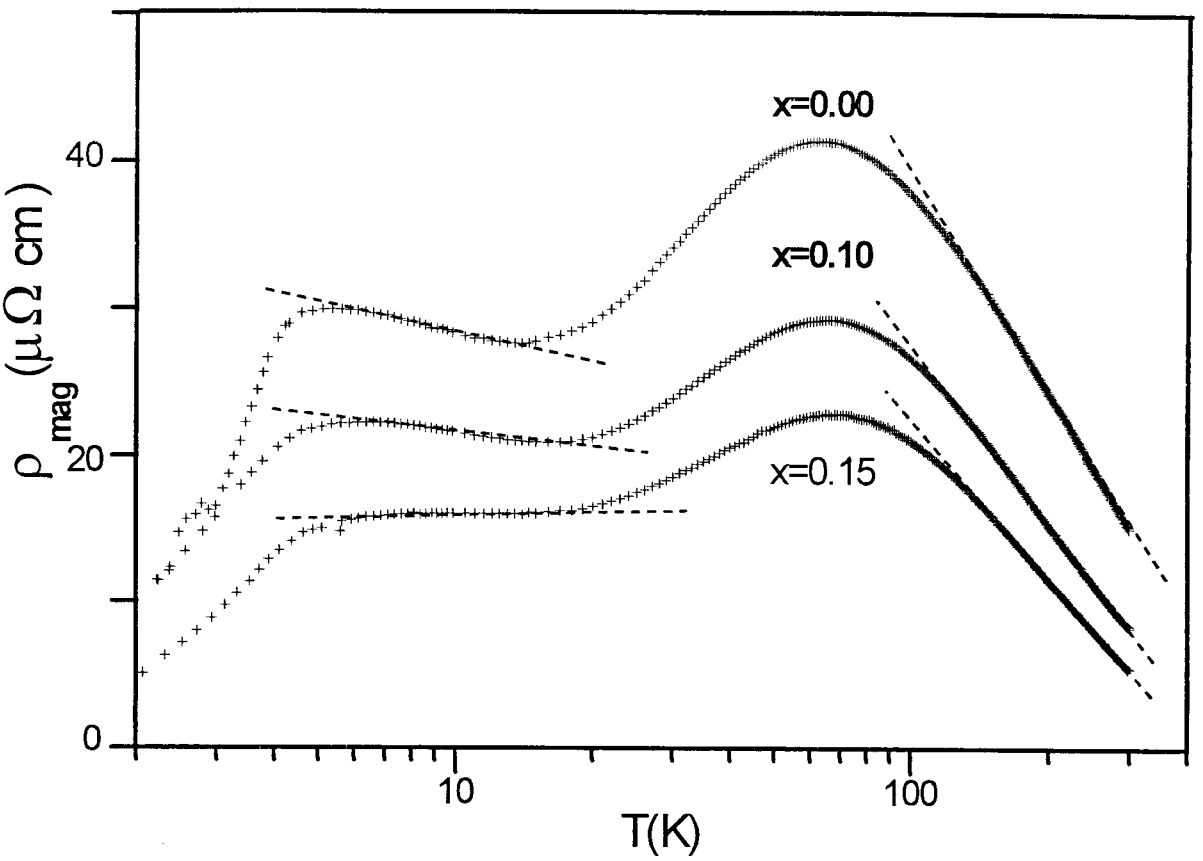

Figure 5

TAB. I

x	a (Å)	c (Å)	c/a	a (Å) cub	T_N (K)	Refs.
1.00	4.285	4.337	1.012		9.5	9,10
0.90	4.297	4.334	1.009		9.5	9,6
0.50	4.378	4.329	0.989		6.0	12
0.40	4.370	4.290	0.982		4.3	this work
0.30	4.390	4.250	0.968		4.0	"
0.20	4.380	4.260	0.973	8.040	3.7	"
0.15				8.040	3.8	"
0.10				8.048	4.1	"
0.05				8.051	4.2	"
0.00				8.064	4.2	"

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