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# Competition between RKKY and Kondo interactions in $CeSi_{2-x}Ga_x$

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**Abstract.** Electrical resistivity measurements on compounds belonging to $CeSi_{2-x}Ga_x$ ($0.7 \leqslant x \leqslant 1.3$) have been performed. The samples in this concentration range show a smooth crossover from a RKKY dominated ground state to a state where the single site Kondo interactions dominate. The variation of Kondo temperature and the magnetic ordering temperature with Ga concentration agree well with the Kondo lattice model. An attempt has also been made to calculate the resistivity curves in these compounds from our quasielastic neutron line width data and using Freimuth's model modified by Garde and Ray. Our results remove an important deficiency in the latter model concerning the motion of the f level relative to the Fermi level.

## 1. Introduction

The competition between the single site Kondo interactions leading to quenching of magnetic moment and the magnetic exchange interactions resulting in a magnetically ordered ground state has been attracting the attention of researchers for more than a decade [1, 2]. The two mechanisms compete with each other with their respective energy scales *viz.* $k_BT_K$, where $T_K$ is the Kondo temperature, and $k_BT_R$, where $T_R$ is the temperature associated with RKKY ordering. The two characteristic temperatures can be expressed in terms of $|JN(E_F)|$, by $T_K \sim \exp(-1/|JN(E_F)|)$ and $T_R \sim |JN(E_F)|^2$, where $N(E_F)$ is the electronic density of states at the Fermi level and $J$ is the exchange interaction between the f and conduction electrons. Hence, for small values of $|JN(E_F)|$, $T_R > T_K$ and magnetic interactions dominate over Kondo interactions, while for larger values of $|JN(E_F)|$, $T_K > T_R$. This changes the behaviour of the rare earth (Ce) from magnetic 4f metal to magnetic concentrated Kondo system (CKS) and from magnetic CKS to non-magnetic CKS.

The system $CeSi_{2-x}Ga_x$ exhibits various types of ground state as $x$ is varied from 0 to 2. In the region $0 \leqslant x \leqslant 1.3$ the compounds crystallize in the $\alpha$-$ThSi_2$ type tetragonal structure ($I4_1/amd$) with a small two phase region in the concentration range $0.2 \leqslant x \leqslant 0.5$ [3]. The compounds with $0.7 \leqslant x \leqslant 1.3$ order ferromagnetically at low temperature [4]. Beyond $x = 1.4$ the compounds have the $AlB_2$ type hexagonal structure ($P6/mmm$) with $x = 1.4$ being ferromagnetically ordered [5] while compounds with $x \geqslant 1.5$ have antiferromagnetic ground state [6]. $CeGa_2$ shows multiple magnetic transitions between 11 and 8.5 K [7].

In this paper, we report measurements on the electrical resistivity in three compounds belonging to the series $CeSi_{2-x}Ga_x$ with $x = 0.7$, 1.0 and 1.3. Resistivity measurements

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have been reported earlier [4]. In this work we have carried out the measurements on the same samples on which we have performed inelastic neutron scattering [8] with the aim to study the resistivity behaviour in these compounds in conjunction with our neutron scattering data. Resistivity apart from being sensitive to the competition between the RKKY and Kondo interactions is also sensitive to the variations in $N(E_F)$ at the Fermi level which in turn depends on the hybridization of the 4f and conduction electrons. We have also calculated the resistivity for all three compositions using our neutron scattering linewidths [8] based on the Freimuth model [9] which has recently been modified by Garde and Ray [10].

## 2. Experiment

The alloys of $CeSi_{2-x}Ga_x$ having compositions $x = 0.7$, 1.0, 1.3 were prepared by arc melting stoichiometric amounts of Ce, Si and Ga in an argon atmosphere. All the buttons were flipped over and remelted at least four to five times to ensure good homogeneity. The phase purity of the samples was confirmed by x-ray diffraction. The lattice parameters determined from the x-ray patterns agree well with those reported earlier [3] and along with the magnetic ordering temperature of these compounds are listed in table 1.

**Table 1.** Lattice parameters and magnetic ordering temperature for $CeSi_{2-x}Ga_x$.

| Concentration $x$ | $a$ (Å) | $c$ (Å) | $c/a$ | $V$ (Å$^3$) | $T_C$ (K) |
|---|---|---|---|---|---|
| 0.7 | 4.237(4) | 14.10(2) | 3.328 | 253.1 | 12 |
| 1.0 | 4.239(7) | 14.14(3) | 3.336 | 254.1 | 7 |
| 1.3 | 4.244(2) | 14.15(3) | 3.334 | 254.9 | 2 |

The electrical resistivity was measured by the standard four probe technique [11]. For the measurements the buttons were cut into thin rectangular plates of approximate size $10 \times 4 \times 1$ mm$^3$. The data were recorded from 4.2 K to 300 K, with current being applied in both directions to eliminate thermovoltage due to the temperature gradient within the connection wires. The control accuracy in temperature was better than 10 mK. Magnetic resistivity, $\rho_m$, was obtained by subtracting the resistivity of LaSiGa from all the Ce compounds studied here assuming the validity of the Mattheiessen rule.

## 3. Results and discussion

In figure 1 we plot the normalized resistivity ($\rho/\rho_{300}$) against temperature for the three compounds. It is to be noted that the presence of a sharp cusp in the $\rho$ versus $T$ data for $x = 0.7$ corresponds directly to the onset of magnetic order. As the Ga concentration increases the sharp cusp slowly disappears with a small hump being visible in $x = 1.0$ data. In the case of $x = 1.3$ no such hump is seen. The change in the shape of the resistivity curve with $x$ indicates a transition from a region of RKKY dominance to a region where the Kondo interactions dominate. A similar change of behaviour has also been observed by Lee *et al* [12] for the $CeSi_x$ system.

In figure 2 we have plotted the magnetic resistivity against temperature for all three compounds. The concentrated Kondo systems are generally characterized with two Kondo temperatures known as the high Kondo temperature ($T_K^h$) where the entire $J = 5/2$ multiplet

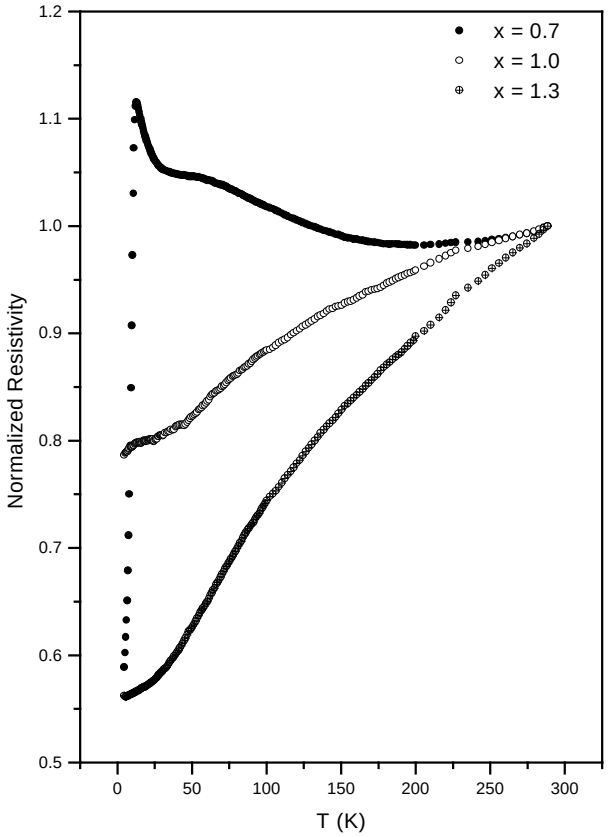


**Figure 1.** Normalized resistivity ($\rho/\rho_{300}$) of $CeSi_{2-x}Ga_x$, $x = 0.7$, 1.0 and 1.3.

**Table 2.** The two Kondo temperatures, $T_K^h$ and $T_K^l$ and the crystal field excitation energies and the Kondo temperature estimated from neutron scattering linewidths, $T_K(QE)$ for $CeSi_{2-x}Ga_x$.

| $x$ | $T_K^h$ (K) | $T_K^l$ (K) | $\Delta_1$ (K) | $\Delta_2$ (K) | $T_K(QE)$ (K) |
|---|---|---|---|---|---|
| 0.7 | 60 | 9.6 | 150 | 150 | 10 |
| 1.0 | 98 | 18 | 174 | 310 | 17 |
| 1.3 | 100 | 27 | 127 | 232 | 29 |

participates in the on-site scattering and the low Kondo temperature ($T_K^l$) where the each crystal field split level participates individually in the on-site scattering [13]. $T_K^h$ and $T_K^l$ for these compounds were calculated using the relation, $T_K^h = (T_K^l \Delta_1 \Delta_2)^{1/3}$: here $\Delta_1$ and $\Delta_2$ are the crystal field excitation energies. From this relation $T_K^l$ was estimated by using the values of $\Delta_1$ and $\Delta_2$ from our neutron scattering results [8] on these compounds, and $T_K^h$ was taken to be the temperature at which the magnetic resistivity shows a maximum (see arrow marks in figure 2). Table 2 gives the values of $T_K^h$, $T_K^l$, $\Delta_1$ and $\Delta_2$ along with Kondo temperature estimated from neutron scattering linewidths taken from [8] for these compounds. In figure 3 we have plotted $T_C$ and $T_K^l$ versus $x$. It is interesting to see that the Kondo temperatures estimated from neutron scattering linewidths and $T_K^l$ not only

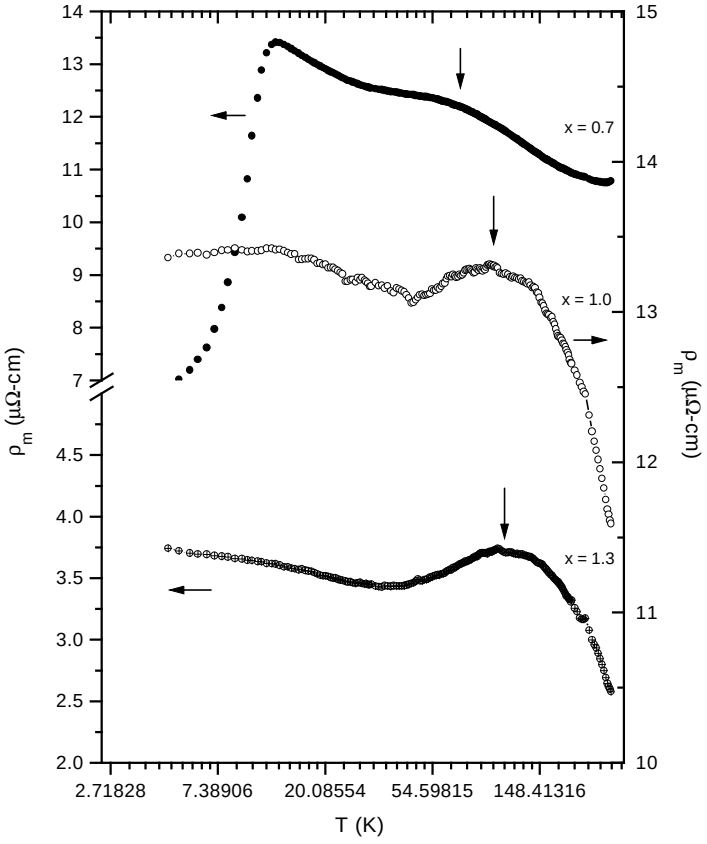


**Figure 2.** Magnetic resistivity ($\rho_m$) of $CeSi_{2-x}Ga_x$, $x = 0.7$, 1.0 and 1.3.

have a similar behaviour with Ga concentration but are closely similar in magnitudes. This information is quite important in knowing the exact Kondo temperatures in such ferromagnetic alloys which are quite rare among cerium Kondo compounds. The behaviour of $T_C$ and $T_K$ in this figure is consistent with the theoretical treatment of the Kondo lattice [14–16]. The theoretical treatment [14–16] predicts a systematic dependence of $T_C$ and $T_K$ on the interaction strength $|JN(E_F)|$ which closely resembles the behaviour shown in figure 4. The similarity between the experimental data and the theoretical predictions indicates that the interaction strength varies with concentration of Ga. This can be shown to be due to increasing hybridization between the 4f and conduction electrons with increasing Ga concentration. This is in agreement with the prediction of the Kondo lattice model [14, 15]. Figure 4 gives the well known qualitative picture of the competition between Kondo effect and the magnetic ordering. Here $T_K$ represents Kondo temperature for a single impurity, $T_{RKKY}$ the temperature associated with RKKY interactions and the magnetic ordering temperature is shown by $T_C$. $T_C$ firstly increases with increasing $|JN(E_F)|$, then passes through a maximum and tends to zero at a critical value, $|JN(E_F)|_c$. In the case of $CeSi_{1.3}Ga_{0.7}$, $T_C$ is higher than $T_K^l$; hence the RKKY interactions exceed the local 4f conduction electron scattering and magnetic order develops. The heat capacity data [4] also show a sharp anomaly at the magnetic transition temperature indicating the clear dominance of RKKY interactions over the Kondo interactions. The magnetic entropy in this compound

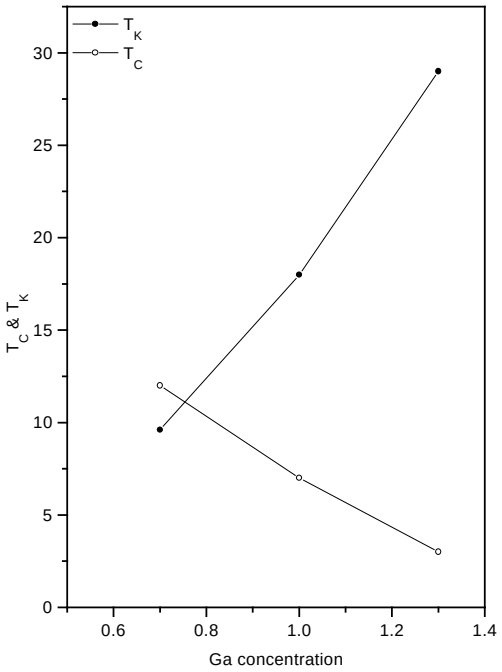


**Figure 3.** Plot showing the variation of $T_C$ and $T_K$ versus Ga concentration in $CeSi_{2-x}Ga_x$, $x = 0.7$, 1.0 and 1.3.

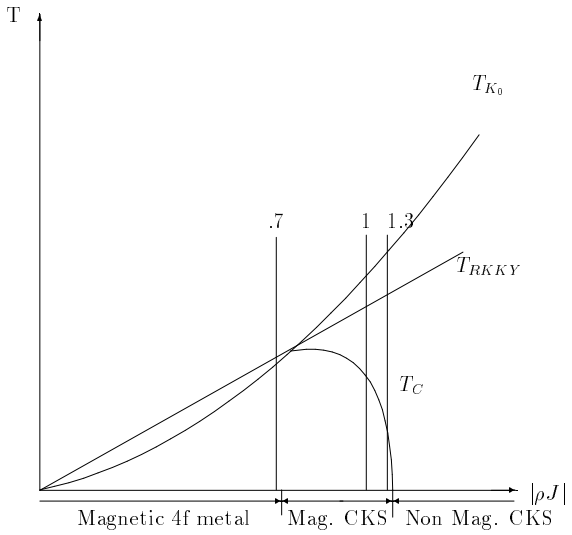


**Figure 4.** Theoretical plot of $T_C$ and $T_K$ variation with $|JN(E_F)|$ in cerium based systems.

is very close to the expected $R \ln 2$ value. Therefore this compound is placed in the magnetic 4f metal region (see figure 4).

For $x = 1.0$, although $T_K^l > T_C$ the intersite magnetic interactions have sufficient strength to cause a magnetically ordered ground state. All the same significant Kondo

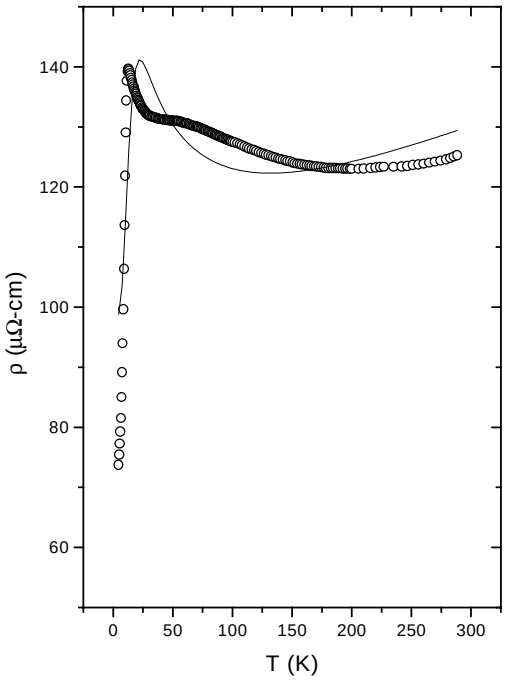


**Figure 5.** Plot of calculated and experimental resistivity in $CeSi_{1.3}Ga_{0.7}$.

**Table 3.** Various parameters obtained from simulation studies.

| $x$ | $\rho_0$ ($\mu\Omega$ cm) | $\alpha$ ($\mu\Omega$ cm K$^{-1}$) | $E$ ($\mu\Omega$ cm K) | $A$ (K) | $B$ (K) | $T_m$ (K) |
|---|---|---|---|---|---|---|
| 0.7 | 206.45 | 0.43 | 2766.43 | −6.725 | 36.847 | 150 |
| 1.0 | 150.67 | 0.43 | 1469.37 | −60.59 | 153.04 | 174 |
| 1.3 | 122.46 | 0.32 | 885.531 | −72.048 | 133.94 | 127 |

screening by 4f exchange scattering is present in the ordered state. It is precisely due to this reason we have placed this compound in the 'magnetic concentrated Kondo system' region at a point of magnetic instability.

For $x = 1.3$, $T_K^l \gg T_C$ and there is a significant decrease in the ordering temperature, $T_C = 2$ K. This is in accordance with the Doniach diagram. Here although the Kondo interactions have strengthened compared to the previous compound, RKKY interactions still succeed in establishing magnetic order. Such materials are often called 'ferromagnetic dense Kondo systems' and hence its position in figure 4. From this discussion it is clear that in $CeSi_{2-x}Ga_x$ especially in the region of $0.7 \leqslant x \leqslant 1.3$ there is a crossover in Ce behaviour from a magnetic 4f metal-type behaviour to a magnetically ordered concentrated Kondo system.

### *3.1. Resistivity and neutron scattering linewidths*

In this section we present the calculation of the resistivity in all the three compounds using a phenomenological model originally proposed by Freimuth [9] and recently modified by

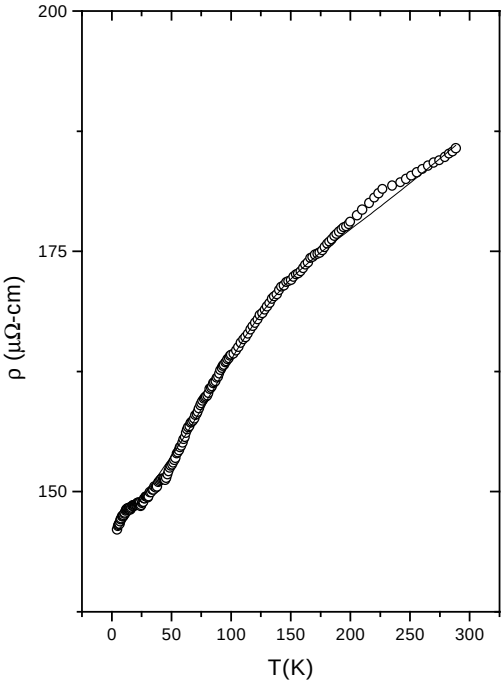


**Figure 6.** Plot of calculated and experimental resistivity in $CeSi_{1.0}Ga_{1.0}$.

Garde and Ray [10]. In the original paper [9] the author has convincingly shown that there exists a correlation between the temperature dependence of the quasielastic linewidth obtained from neutron scattering studies ($\Gamma_{QE}$) and the occurrence of resistivity maxima in Ce compounds. He argued that the major contribution to $\rho$ arises from the scattering of electrons between the broad conduction band and the narrow Lorentzian shaped 4f band, which is proportional to an effective density of f states at the Fermi level $N_f(E_F)$ which has been expressed in the following form [9]

$$N_f(E_F) = \frac{W}{(T_0^2 + W^2)} \tag{1}$$

where $W = T_f \exp(-T_f/T)$ and $T_f$ is a temperature dependent parameter equated to $\Gamma_{QE}$ and $k_B T_0 = (E_F - E_f)$, where $E_f$ is the energy corresponding to the centre of gravity of the 4f peak in the density of states at $E_F$. Thus $T_0$ can be either positive or negative depending on whether $E_f$ is less than or greater than $E_F$ respectively. In the paper [9], the parameter $T_0$ was taken to be temperature independent and positive to calculate the resistivity and thermopower in case of mixed valent compounds. In order to explain the change of sign in thermopower in case of concentrated Kondo systems (CKSs), Garde and Ray [10] introduced temperature dependence for $T_0$ of the form $T_0 = A + B \exp(-T_m/T)$, where $A$, $B$ and $T_m$ are constants for a given alloy. For $T < T_m$, $T_0 \sim A$ and this corresponds to Kondo scattering from Ce ions in their crystal field split ground state. At temperatures $T \gg T_m$, $T_0 \sim A + B$ which means that Kondo scattering now occurs from all the crystal field split levels. The exponential term, therefore can be taken to represent gradual filling up crystal field split levels. Therefore we propose that $T_m$ could be taken to be equal to the crystal splitting energy of the $J = 5/2$ ground state. However, one can no longer consider the definition $k_B T_0 = (E_F - E_f)$ to be still valid, because it turns out that in the case of

mixed valent compounds the 4f level is well below $E_F$ while in the case of Kondo lattice compounds the 4f level moves from below $E_F$ at high temperatures to above $E_F$ at low temperatures and this cannot be true. Hence we feel that $T_0 = A + B\exp(-T_m/T)$ only represents the movement of 4f level as a function of temperature with respect to Fermi level. Here the absolute value of $T_0$ is not important but only its behaviour as a function of temperature. Hence in the case of mixed valent compounds wherein the 4f level is hybridized with Fermi level, $T_0$ does not show any temperature dependence while in case of Kondo compounds it does as the 4f level moves closer to Fermi level with decreasing temperatures.

The resistivity of the Kondo systems can be calculated using,

$$\rho = \rho_0 + \alpha T + E\frac{W}{T_0^2 + W^2} \tag{2}$$

where $\rho_0$ is the temperature independent residual resistivity term, $\alpha$ is a constant which typifies the strength of the nonmagnetic phonon term and $E$ is another constant which relates to the strength of the 4f band term. In our fitting procedure $\rho_0$ was derived from the low temperature (4 K $< T <$ 9 K) data and $\alpha$ was obtained from the slope of the high temperature (250 K $< T <$ 300 K) data in respective compounds. $W$ was estimated from $\Gamma_{QE}$ taken from [7]. Since thermopower data is not available in these compounds the constant $A$ and $B$ were chosen as fitting parameters along with $E$. The parameter $T_m$ was taken to be equal to $\Delta_1$. The values of the different parameters obtained from the fitting are given in table 3 and the experimental data along with fitted curves are shown in figures 5 to 7 for $x = 0.7$, 1.0 and 1.3 respectively. The fitting except for $x = 0.7$ is quite good. In the case of $x = 0.7$ the fitted curve reproduces the general trend of the experimental curve

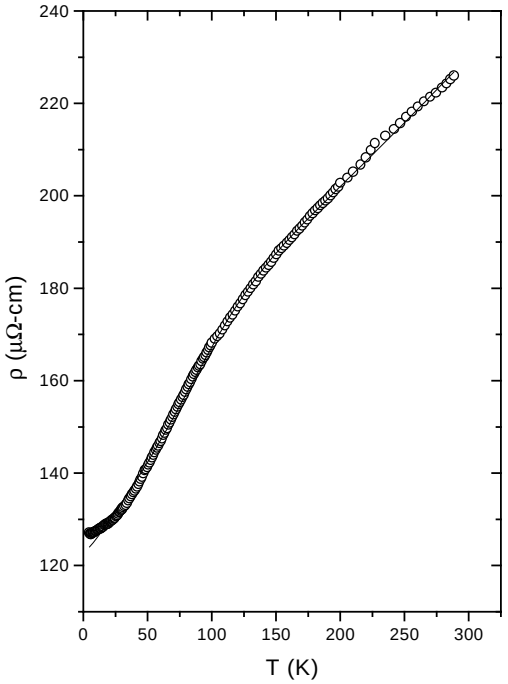


**Figure 7.** Plot of calculated and experimental resistivity in $CeSi_{0.7}Ga_{1.3}$.

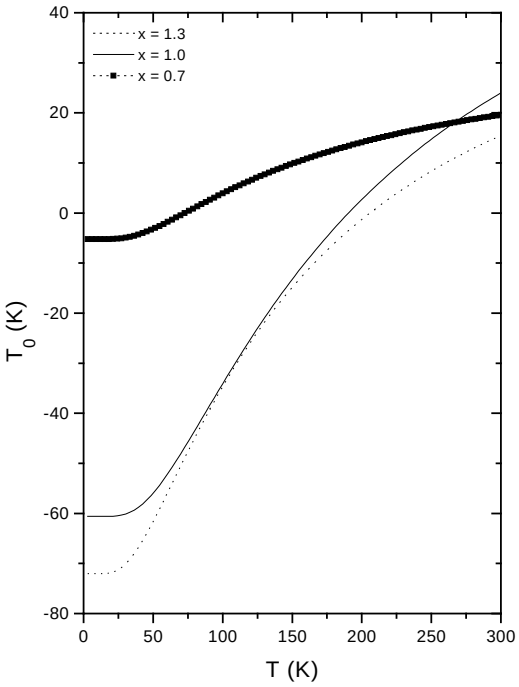


**Figure 8.** Plot of $T_0$ with $T$ (K) in $CeSi_{2-x}Ga_x$, $x = 0.7$, 1.0 and 1.3.

with some reliable parameters. Plotting $T_0$ as a function of temperature in figure 8 we see a large variation for $x = 1.0$ and 1.3 samples, whereas for $x = 0.7$ the variation in $T_0$ is restricted to a narrow region compared to that for the other two samples. Now considering the above argument of the variation of $T_0$ we feel this phenomenologically represents the localization of the 4f level in the case of compounds wherein RKKY interaction dominates the Kondo interaction. Thermopower measurements, which are planned, are expected to shed more light on this aspect as it is more sensitive to the sign change in $T_0$.

In conclusion, we have carried out resistivity measurements on $CeSi_{2-x}Ga_x$, $x = 0.7$, 1.0 and 1.3. This series in this concentration region exhibits a crossover from magnetic 4f metal-type behaviour of the Ce ion to a magnetically ordered dense Kondo system. Phenomenological calculation of resistivity behaviour also supports this view.

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