Antiferromagnetism in $Ce_{1-x}La_xAl_2Ga_2$ and $Ce_{1-y}Y_yAl_2Ga_2$ Kondo-lattice systems

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We report here resistivity ρ and thermoelectric-power S measurements between 1.7 and 300 K on $Ce_{1-x}La_xAl_2Ga_2$ and $Ce_{1-y}Y_yAl_2Ga_2$ Kondo-lattice alloys. By variation of x and y, there is a clear evolution of the Kondo-like resistivity minimum, which is not seen in the parent alloy ($x=0$). With both La and Y substitutions, the antiferromagnetic transition temperature T_N is found to be depressed. It is suggested that this depression by a variation of x can be explained solely on the basis of the weakening of the intersite Ruderman-Kittel-Kasuya-Yosida interaction, whereas with a variation of y, the T_N depression results from an interplay of both single-site and intersite effects.

Among the Ce-based heavy-fermion (HF) compounds of the CeT_2Si_2 (T=Au, Pd, Rh, Ag) and CeM_2Ge_2 (M=Ag, Au, Pd) types,[1-3] several of them show antiferromagnetic (AF) ordering at temperatures $T_N < 15$ K. It is well known that this magnetic ground state is a result of a subtle interplay between single-site (Kondo) and long-range effects arising from intersite Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between the Ce ions. The characteristic temperature scales of the above two interactions are designated by T_K and T_{RKKY}, respectively. The precise nature of this interplay determines the shape of the ρ and S curves at low temperatures.[4-6] The presence of the Kondo effect is usually manifested by the occurrence of ρ_{min} at T_{min} with the magnetic resistivity increasing ($\partial\rho/\partial T<0$) for $T<T_{min}$. Eventually, the ρ curve passes through a maximum at the ordering temperature T_N. At still lower temperatures, the HF coherent phase and the AF phase is found to coexist in many of these systems. Measurements of S on the other hand show a negative minimum at a temperature close to T_{min} reflecting the onset of Kondo behavior. In several such compounds, S changes sign and becomes positive at still lower temperatures. This crossover of sign is treated as an evidence for the formation of the coherent HF bands.[7-10]

$CeAl_2Ga_2$ was found to exhibit the same crystal structure ($ThCr_2Si_2$ type) as of the above class of compounds and it has been of recent interest[11-13] to see how physical properties of this new compound compared with them. Neutron studies[11,12] on single and polycrystals of $CeAl_2Ga_2$ reveal the presence of AF state at $T_N=8.9$ K. The magnetic susceptibility was found to be anisotropic. Departure from Curie-Weiss law occurs below 60 K. At high temperatures, moment on the Ce atoms was found to be close to the Ce^{3+} free-ion value.[13] Like $CeRu_2Si_2$, Kondo effect features (like ρ_{min}) were not very pronounced, possibly due to the presence of magnetic correlation effects above T_N. A λ-type anomaly is seen in the specific heat data[11] at the same temperature where ρ_{max} occurs. $GdAl_2Ga_2$ does not order magnetically for $T>20$ K. On the basis of de Gennes scaling,[13] T_N of $CeAl_2Ga_2$ should have been around 0.1 K or less. Therefore the value of T_N for $CeAl_2Ga_2$ is anomalously high.

In view of the complex magnetic behavior of the Kondo-lattice (KL) compound $CeAl_2Ga_2$, it is felt that it would be interesting to study the following two systems, namely, $Ce_{1-x}La_xAl_2Ga_2$ (x=0, 0.05, 0.1, 0.2, 0.5, 0.8) and $Ce_{1-y}Y_yAl_2Ga_2$ (y=0.05, 0.15, 0.30). The variation of x and y in these two systems would provide a means to alter the exchange parameter J, arising out of the interaction between the Ce $4f$ electron and the conduction electrons. This would enable us to control the relative strengths of T_K and T_{RKKY} and study the variation of T_N with J. All the alloys were prepared in an arc furnace by repeated melting in flowing argon. For lower impurity concentrations, the x rays of these systems showed a single phase of the $ThCr_2Si_2$ type. For $x\geq 0.5$ and $y\geq 0.4$, evidence of extra lines in the x-ray patterns were observed indicating the growth of a second phase. The values of the lattice parameters a and c for $CeAl_2Ga_2$ were close to earlier reported values.[13,14] With an increase of x, the value of a increased smoothly, whereas the value of c decreased keeping the increase of cell volume V to about 1% at $x=0.8$ over the parent compound ($x=0$). On the other hand, with an increase of y, both a and c parameters decreased relatively rapidly resulting in 1% change in V at $y=0.3$. Thus the effect of negative and positive pressure with La and Y substitution, respectively, is seen clearly reflected in unit cell volume V, the effect being more marked in the latter case. As the samples were brittle, definite geometric shapes were difficult to obtain. Measured resistivity has therefore been expressed in arbitrary units.

In the ρ studies (Figs. 1 and 2) the low-temperature magnetic transition is reflected in ρ_{max} below which ρ drops rapidly. ρ_{max} is identified with T_N. For $x=0$, $T_N=9.2$ K (Fig. 3), in close agreement with a previous study.[11] The most interesting feature is the gradual decrease of T_N with La and Y substitution (see inset of Fig. 1). For $x=0.8$, T_N drops to 2.5 K (Fig. 3) whereas for $y=0.3$, $T_N=5.3$ K (Fig. 4). The value of T_N was not found to be sensitive to annealing effects (800 °C for 10 days) which was measured for samples with $x\geq 0.5$. An interesting feature is that ρ_{min} which is not clearly ob-

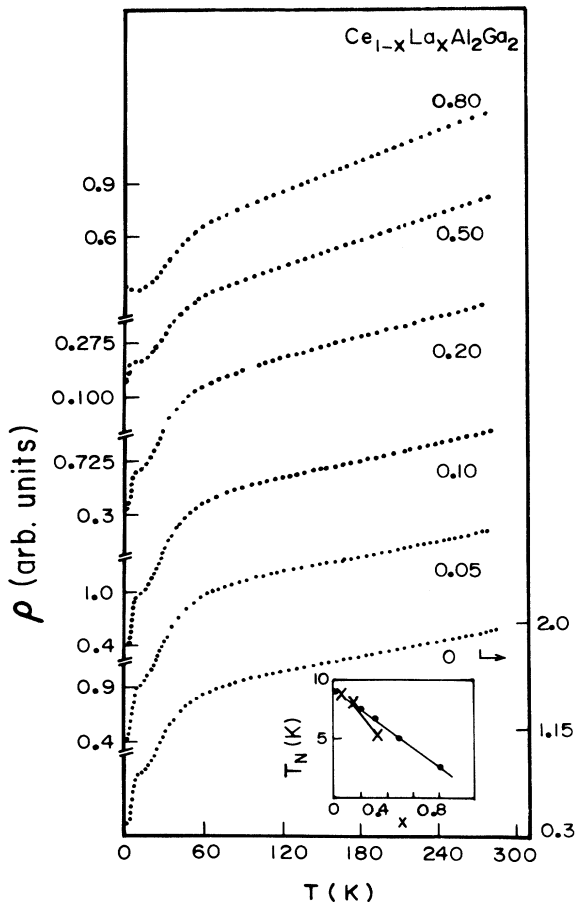

FIG. 1. ρ vs T for $Ce_{1-x}La_xAl_2Ga_2$ alloys. The inset shows the linear variation of T_N with La(x) and Y(y) substitutions. Dots represent T_N for different x values and crosses represent T_N for different y values.

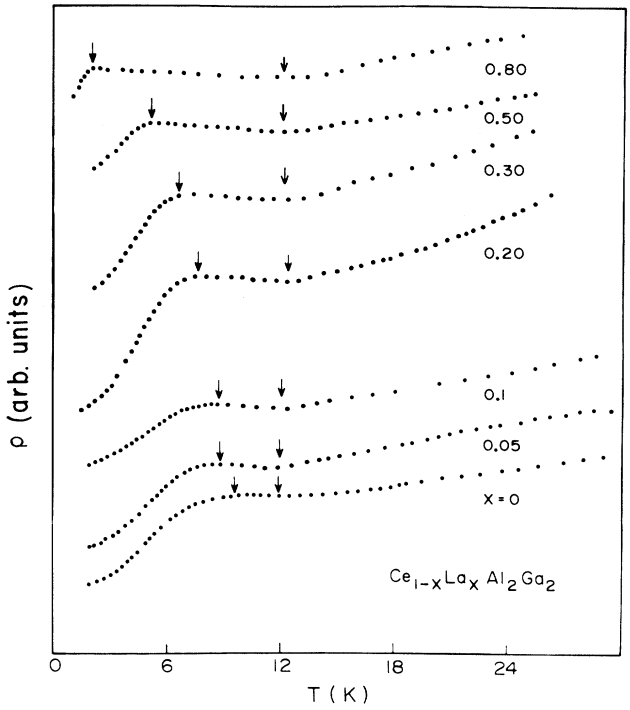

FIG. 3. Low-temperature plots of ρ vs T for $Ce_{1-x}La_xAl_2Ga_2$ alloys. Positions of ρ_{min} are shown by the arrows. We note that ρ_{min} is very shallow for $x=0$, but becomes relatively pronounced for increasing values of x. The arrows at still lower temperatures indicate T_N at ρ_{max}.

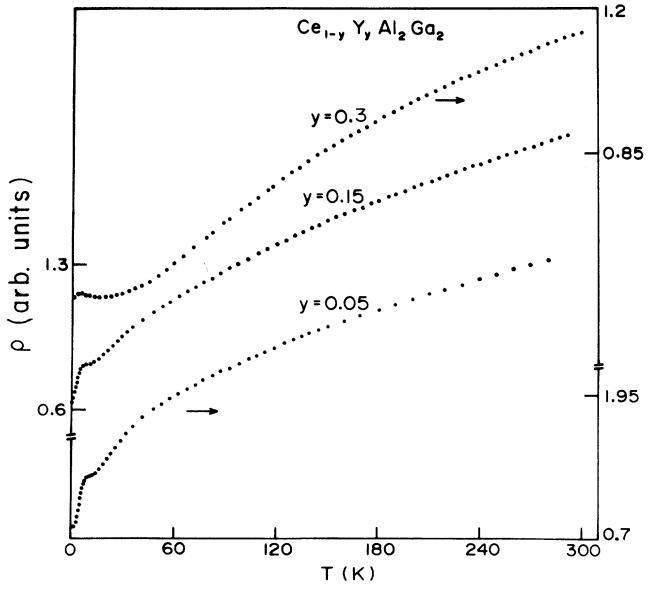

FIG. 2. ρ vs T curves for $Ce_{1-y}Y_yAl_2Ga_2$ alloys.

served for $x=0$ (Fig. 3), begins to appear as x or y is increased. From our studies, it is found that ρ_{min} is more pronounced with variation of y than with x (Figs. 3 and 4). It was inferred[11] that the near absence of ρ_{min} in pure $CeAl_2Ga_2$ ($x=0$) is most probably related to magnetic correlation effects as revealed from the occurrence of re-

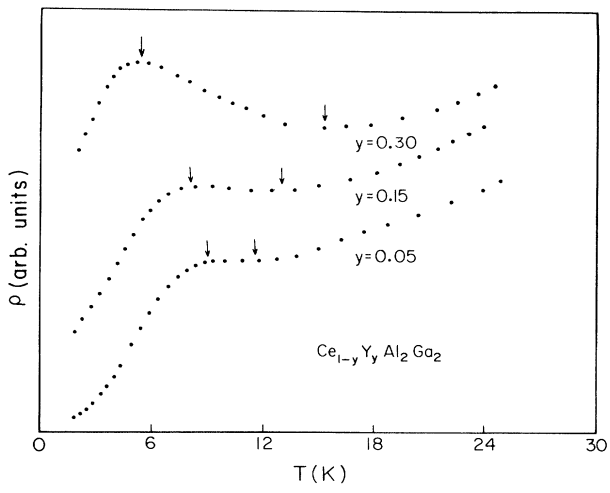

FIG. 4. Low-temperature plots of ρ vs T for $Ce_{1-y}Y_yAl_2Ga_2$ alloys. Positions of ρ_{min} are shown by the arrows. The arrows at still lower temperatures indicate T_N at ρ_{max}.

duced entropy at T_N from specific heat studies. These magnetic correlations could give rise to extra resistivity thus suppressing the manifestation of the Kondo minimum. It could therefore mean that with La and Y substitutions, the effects of magnetic correlations become weaker thus enabling Kondo effect to manifest. The variation of T_{min} with x and y are as shown in Figs. 3 and 4. T_{min} is found to be constant around 12 K for all values of x (Fig. 3) whereas it is found to vary with y. T_{min} is evaluated to be 11.5, 13, and 15 K for $y=0.05$, 0.15, and 0.3, respectively (Fig. 4). We note that though the existence of ρ_{min} and the existence of the temperature regime $\partial\rho/\partial T<0$ (for $T<T_{min}$) indicate the presence of a Kondo anomaly, an unambiguous determination of T_K from our ρ data is difficult. In the case of dilute magnetic impurities in a nonmagnetic host,[15] it is found that though T_K could occur for $T<T_{min}$, such an estimate may not always be true under the influence of appreciable phonon background and contributions from low-lying crystal-field (CF) states as is the case in our systems. However, as our studies reveal the presence of AF ordering in all the alloys, it is likely that T_K cannot be very large compared to T_N, as then a nonmagnetic ground state would have been preferred. Our S studies also throw light on possible T_K values for these alloys. It is well known that S is sensitive to scattering from Ce ions and shows a pronounced negative minimum at T_m (close to T_{min}) due to Kondo effect. It has been empirically deduced[8,16] that $T_m=2T_K$, which is found to hold good (within a factor of 2) for nonmagnetic KL compounds[8] like $CeAl_3$ and $CeCu_2Si_2$ and magnetic compounds[8,16–18] like $CePb_3$ and $CeAl_2$. Using this formula for our alloys we find that T_K is constant at 10 K for all values of x (Fig. 5) whereas for $y\leq 0.15$, $T_K=11$ K increasing to 15 K for $y=0.3$ (Fig. 6). We thus find from these estimates that T_K is comparable in magnitude to T_N. Further, our S studies also show signatures of Kondo-like scattering in the presence of CF levels at higher temperatures. This CF derived maximum at T_M is clearly exhibited for $T>50$ K. $T_M=75$ K for $x\leq 0.2$ and increases to 85 K for $x>0.2$ (Fig. 5), whereas $T_M=70$ K for $y\leq 0.15$ and 90 K for $y=0.3$ (Fig. 6). Such evidence of CF excitations are not seen in the ρ plots (Figs. 1 and 2) due to the influence of a large phonon background. This effect is pronounced and exhibits[11] a peak around 75 K in the ρ curve of $CeAl_2Ga_2$ ($x=0$) only after subtracting the phonon term estimated from the nonmagnetic $LaAl_2Ga_2$ compound.

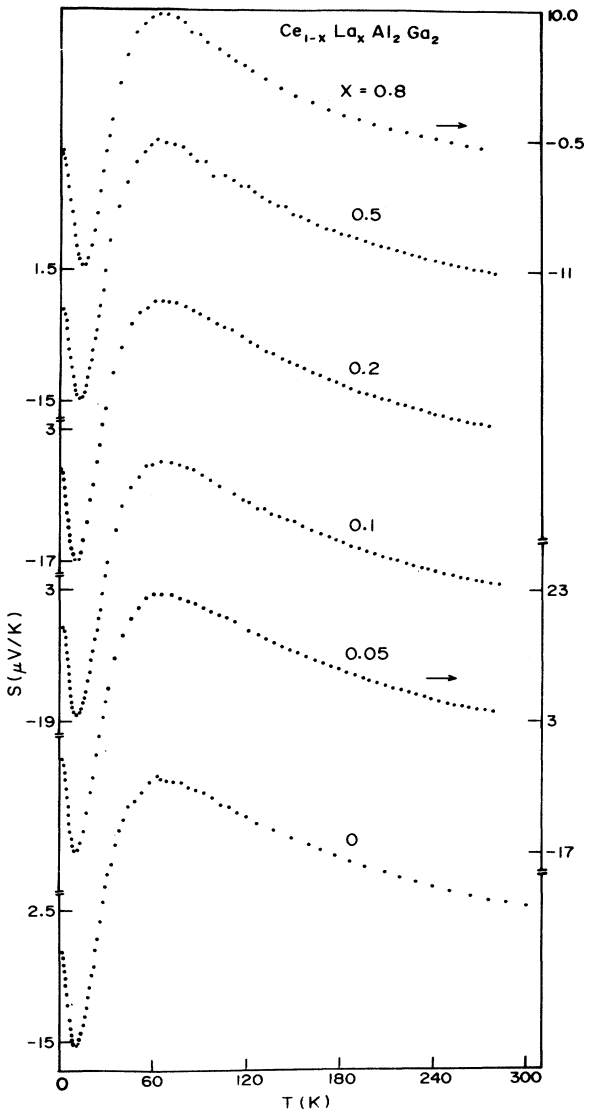

FIG. 5. S vs T curves for $Ce_{1-x}La_xAl_2Ga_2$ alloys.

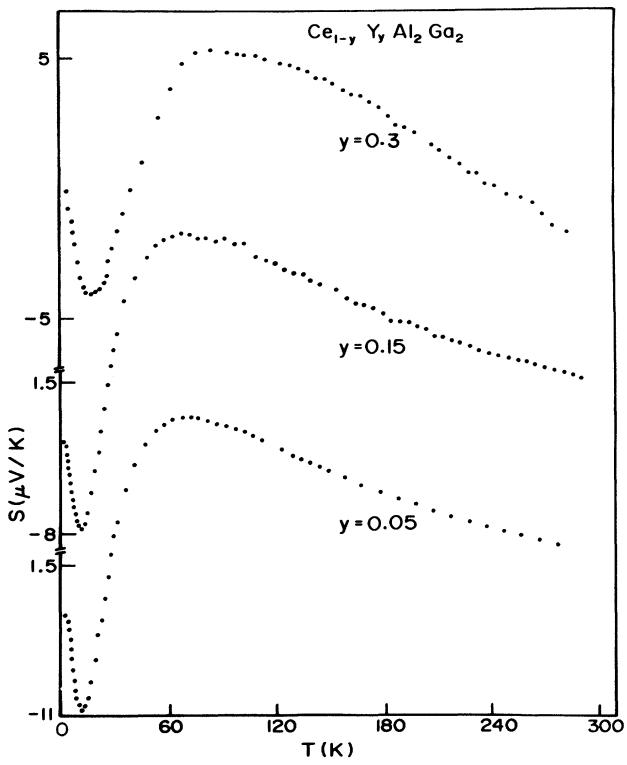

FIG. 6. S vs T curves for $Ce_{1-y}Y_yAl_2Ga_2$ alloys.

We now examine the implications of the T_N variation in our alloys. With variation of x (La substitution), T_N decreases almost linearly down to $x=0.8$ (Fig. 1). Extrapolating to still higher x values, we would find that $T_N \to 0$ as $x \to 1$. The most direct interpretation, for such a linear dependence of T_N depression, could be solely based on the weakening of the RKKY interaction between Ce ions. It is known[19] that the variation of T_{RKKY} is given by $T_{RKKY} \propto cJ^2$, where c is the concentration of Ce atoms. Therefore $T_{RKKY} \to 0$ as $c \to 0$ ($x \to 1$). This shows that the variation of T_N with x is governed by a single parameter T_{RKKY}. On the other hand, the decreases of T_N with increase of y (Y substitution) is relatively faster (Fig. 1). Extrapolating to higher y values, we would find that $T_N \to 0$ for $y < 1$ which is in contrast to the La substituted alloys. Thus the T_N depression with y (Y substitution) cannot be fully explained on the basis of the weakening of T_{RKKY} alone. To understand this we note that the effect of Y substitution (positive pressure on the $y=0$ lattice) leads to relatively larger variation (decrease) of V compared to its increase for the La substituted alloys. As J is inversely proportional[20] to V, it would mean that with increase of y (decrease of V), J would increase. Using the relation $T_K \sim \exp(-1/JN)$ (where N is the density of states at the Fermi level), we find that T_K would also be enhanced with an increase of J. To explain the faster depression of T_N, we therefore propose that apart from T_{RKKY}, T_K also plays an important role in deciding the value of T_N. In the light of Doniach's picture,[21] it would then imply that $CeAl_2Ga_2$ and the Y substituted alloys would lie on the right-hand side of the peak of the $T_N(J)$ phase diagram. However, a firm estimate of T_K by different methods (for both La and Y substituted alloys), would be useful to check the validity of these viewpoints. Further, if such experiments do show the variation of T_K also for the La substituted alloys, then these systems could also be explained in the light of the above $T_N(J)$ diagram. Finally, we note that our S data naively seems to indicate that T_N and T_K for $CeAl_2Ga_2$ are of comparable magnitudes, and if this indeed turns out to be true, the situation would resemble that of $CePd_2Si_2$. In this system, the neutron quasielastic linewidth data[3] estimates $T_K \sim T_N$ at $T=T_N \sim 10$ K. The AF phase is believed to grow in the presence of Kondo fluctuations and it is argued[3] that $CePd_2Si_2$ lies on the peak of the $T_N(J)$ diagram. Further support[22] of this location is attributed to the T_N depression of $CePd_2Si_2$ by La and Y substitutions at the Ce site. In views of such similar trends observed in our systems too, it would be of interest to see if more definitive experiments would support that $CeAl_2Ga_2$ could also be similarly located on the $T_N(J)$ diagram.

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