Kondo-lattice behavior of the interstitial alloys $CeFe_xGe_2$

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The results of magnetic susceptibility, electrical resistivity, magnetoresistance, and heat-capacity measurements on the nonstoichiometric alloys $CeFe_xGe_2$ ($x=0.53$ and 0.63) are reported. Some of the observed properties are typical of (nonmagnetic) Kondo lattices in spite of the nonstoichiometric nature of the chemical composition. These alloys are characterized by a large electronic term in the heat capacity.

It is well known that, in the case of Ce-based Kondo lattices, the electrical resistivity (ρ) exhibits a maximum at a temperature where there is a crossover from the high-temperature incoherent regime to the low-temperature coherent regime due to the periodicity of the lattice.[1] A disruption of the Ce sublattice either by the substitution for Ce (Ref. 2) or by the disorder of the lattice[3] is known to destroy the coherence. During the course of our investigation[4,5] of the nonstoichiometric alloys of the type RT_xZ_2 (R=rare earth, T=transition metals, Z=Ge and Sn) crystallizing in the $CeNiSi_2$-type structure,[6] the temperature dependence of the electrical resistivity (ρ) of the Fe containing ones, $CeFe_xGe_2$ (x=0.53 and 0.63), was found to exhibit a maximum around 25 K with a remarkable fall at low temperatures without any evidence for magnetic ordering. Considering the nonstoichiometric nature of the alloys, it is of interest to explore whether this fall in ρ is a true representation of Kondo coherence. With this primary motivation, we have performed further investigations to probe the low-temperature behavior of these alloys, the results of which are discussed in this article. Among these results, the magnetoresistance behavior appears to be consistent with the existence of coherence effects at low temperatures. Heat-capacity (C) studies establish that these alloys are nonmagnetic heavy fermions with an electronic heat capacity (γ) of the order of 240 mJ/mol K^2 for $x=0.53$ and 320 mJ/mol K^2 for $x=0.63$ at 0.5 K.

These alloys employed in the present investigations are the same as that used in Ref. 4 and the crystallographic details explaining interstitial nature of the alloys were also presented earlier.[4] The samples were also metallographically examined and were found to be homogeneous. Electrical resistivity (ρ) data presented here (1.4–300 K) are the same as those reported earlier;[4] the data down to 0.5 K were also obtained on the same specimens at the Institute for Solid State Physics (ISSP), Tokyo, and the values in the temperature interval 0.5–3 K are found to lie on a straight line for each composition. Heat-capacity (C) measurements (0.5–20 K) were performed by a semiadiabatic heat-pulse method at ISSP; we have also taken C versus T data in the temperature interval 2–100 K employing a calorimeter fabricated recently by Das and Sampathkumaran,[7] and the values of C obtained from both the calorimeters agree well below 20 K. Magnetoresistance up to a magnetic field of 6 T were also measured at selected temperature below 30 K. Magnetic susceptibility (χ) measurements (4.2–300 K) were performed by a Faraday method in a magnetic field of 4 kOe.

For the sake of clarity, we reproduce the results[4] of ρ measurements in Fig. 1. The ρ increases with decreasing temperature. It is interesting to see that the data for $x=0.63$ above 50 K vary nearly linearly with temperature, whereas for $x=0.53$, ρ varies logarithmical typically of that expected for the Kondo effect; the reason for the difference in the temperature dependence between these two compositions is not clear. No attempt is made to extract the $4f$ contribution to ρ, as it is rather difficult to obtain a suitable reference for phonon contribution for such nonstoichiometric alloys. There is a peak at 25 and 15 K (called T_{max}) for $x=0.63$ and 0.53, respectively, followed by a significant drop at lower temperatures. The drop, however, is sharper for $x=0.53$. Earlier magnetization studies have shown[4] that this drop is not related to magnetic ordering. It may be recalled[8] that there has been a controversy regarding the interpretation of T_{max}

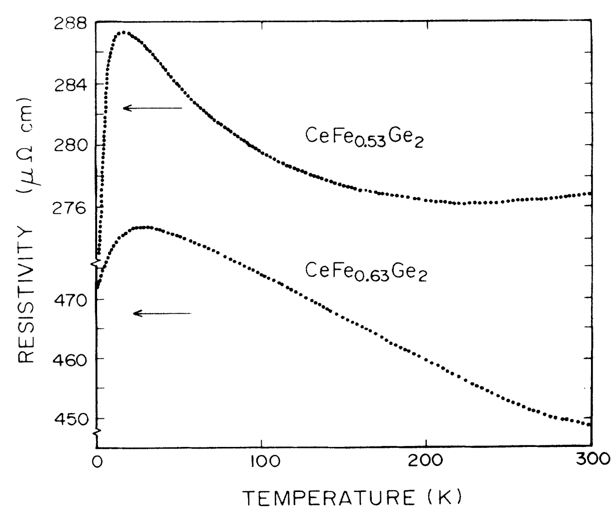

FIG. 1. Electrical resistivity as a function of temperature (1.4–300 K) for the alloys, $CeFe_xGe_2$ ($x=0.53$ and 0.63).

and we have proposed experimental criteria[9] to identify the origin of this peak. According to this criteria, the sensitivity of the low-temperature resistivity behavior to stoichiometry below T_{max} in nonmagnetic Kondo lattices implies that the drop in ρ arises from the coherence among Kondo centers. It may be noted that ρ at T_{max} is larger only by a few percent compared to the 4-K value, in contrast to the situation in conventional translationally invariant Kondo lattices. Hence it may be tempting to attribute this peak to crystal-field peaks; in addition, the absence of T^2 dependence of ρ at low temperatures, expected for Kondo lattices, may at first sight favor this line of interpretation. We offer the following arguments against such interpretations: (i) Since these alloys are disordered, the residual resistivity values are presumably high and hence the ratio of $\rho(T_{max})/\rho(4\text{ K})$ may not be the true representation of the $4f$ contribution to ρ; (ii) If T_{max} represents the crystal-field peak, then one would have expected a Schottky peak in C in the vicinity of T_{max}; the C data (*vide infra*) do not indicate the existence of such an anomaly in both the alloys; (iii) the low temperature T^2 dependence of ρ is expected for translationally invariant Kondo lattices and it is presently not clear how ρ at low temperatures is expected to vary in interstitial alloys, in which the chemical environment for all Ce ions is not the same; therefore, the absence of T^2 dependence of ρ need not be viewed as a finding against coherence behavior in our alloys.

We have also performed magnetoresistance measurements up to 6 T at a few selected temperatures above 4.0 K. We show in Fig. 2 the magnetoresistance, $\Delta\rho/\rho=\{\rho(H)-\rho(0)\}/\rho(0)$ at 4.2 and 10 K for the two alloys. $\Delta\rho/\rho$ is positive at 4.2 K, whereas at 10 K, the values tend to become negative at higher fields. The observed behavior is typical of that expected for Kondo lattices.[1,10] It is generally accepted[1,10] that the coherent scattering among the Kondo scattering normally leads to a drop in the ρ values below T_{max} and the application of the magnetic field (H) destroys this coherence leading to an increase of the absolute values of the resistivity; this explains the positive magnetoresistance at 4.2 K. In the incoherent state, the spin-fluctuation contribution to ρ is suppressed in the presence of H, resulting in a negative magnetoresistance. The values at 10 K below 3 T are too small to draw a definitive conclusion regarding the sign of the magnetoresistance, but at higher fields the values are distinctly negative. We have also taken the data at 20 and 30 K and the values, though small (not shown in the figure) are clearly negative. Thus, the magnetoresistance behavior is consistent with the proposal that there is a transition from a coherent to incoherent scattering state around 10 K in these alloys.

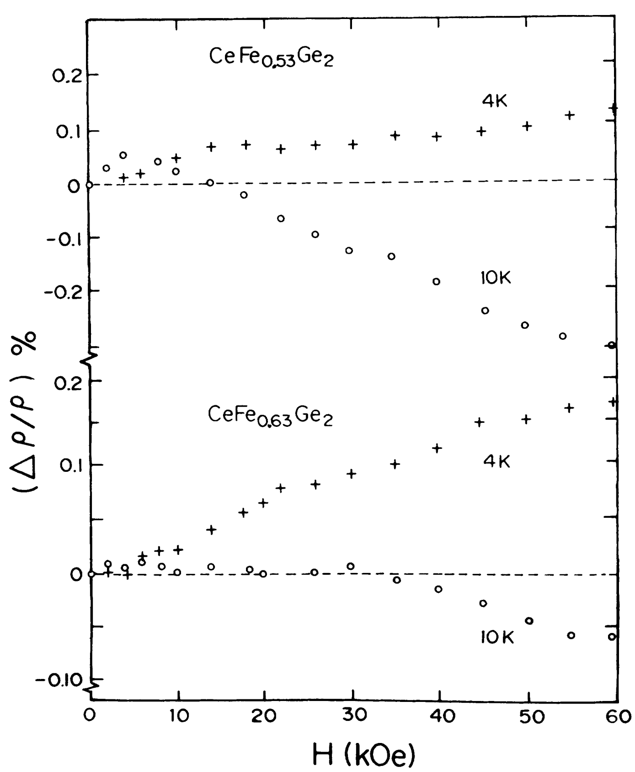

FIG. 2. Magnetoresistance as a function of magnetic field at 4 and 10 K for the two alloys, $CeFe_xGe_2$ ($x=0.53$ and 0.63).

In order to look for crystal-field effects (around 30 K) as well as for other characteristics of Kondo lattices, viz., a large linear term (γ) in the heat capacity (C), we have performed heat-capacity studies in the temperature interval 0.5–100 K. The results are shown in various ways in Figs. 3 and 4. As concluded earlier,[4] there is no distinct feature due to magnetic ordering in the temperature range of investigation. A small peak, however, appears around 6 K (shown in an expanded form in Fig. 4 in the form of C/T versus T^2). It is of interest to explore whether this peak is Kondo-coherence derived,[2] considering that the coherence temperature as inferred from the magnetoresistance data presented above falls in the vicinity of 10 K. Presently, however, we cannot exclude the presence of some impurity phases, presumably about 2–4 % of magnetic Ce oxides, as a possible cause of this feature. It is obvious that there is no distinct peak in the C versus T plot around 30 K. Ideally, one has to subtract

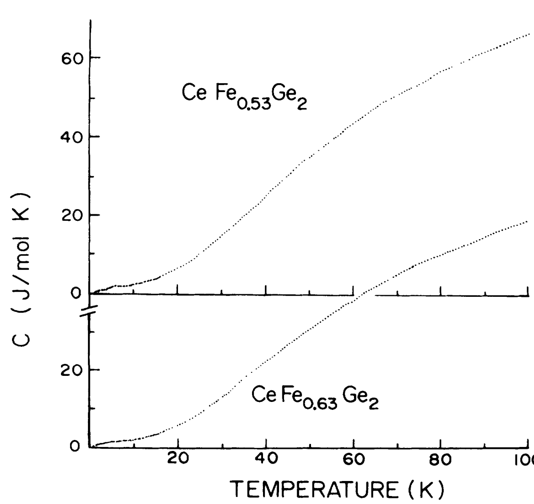

FIG. 3. Heat capacity (C) as a function of temperature for $CeFe_{0.63}Ge_2$ and $CeFe_{0.53}Ge_2$.

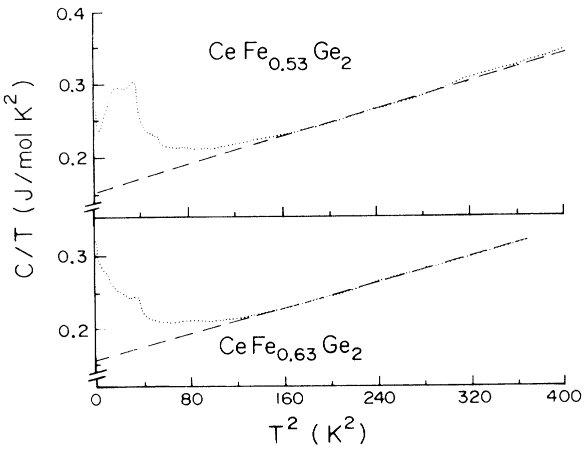

FIG. 4. Heat capacity divided by temperature as a function of temperature square for $CeFe_{0.63}Ge_2$ and $CeFe_{0.53}Ge_2$. The dashed lines are drawn through the data points above 10 K.

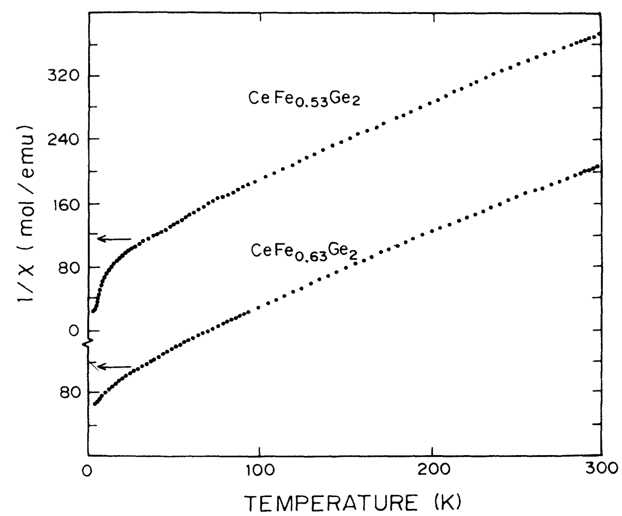

FIG. 5. Inverse susceptibility ($1/\chi$) as a function of temperature for the alloys $CeFe_xGe_2$ ($x=0.53$ and 0.63).

the lattice contribution to C in order to derive the features attributable to $4f$ contribution. As already mentioned, this is a difficult task in such interstitial alloys. Nevertheless, we can state that a Schottky peak around 30 K is not transparent in the raw data. The values of C/T obtained by the extrapolation of the data in the temperature interval 10–20 K to absolute zero (marked by broken line in Fig. 4) is large (160 mJ/mol K^2) and the value increases below 10 K reaching a value of about 320 mJ/mol K^2 for $x=0.63$ and 240 mJ/mol K^2 for $x=0.53$ at 0.5 K. Thus the results suggest that these alloys are heavy fermions.

We compare the magnetic-susceptibility behavior of the two compositions in Fig. 5. The inverse magnetic susceptibility is linear above 100 K, with the magnetic moment slightly above that expected for a tripositive Ce ion ($\mu_{\text{eff}}=2.7-2.8\mu_\beta$) probably due to the polarization of the conduction band. The paramagnetic Curie temperature (θ_p) obtained from the data above 100 K falls in the range $-100--115$ K and the values decrease to about -60 K in the temperature range $+20-100$ K. These values suggest that the single-ion Kondo temperature (T_K) is typically of the order of 15–25 K.[11] This finding suggests that $T_{\max}$ is closely related to T_K in these alloys and, from the behavior of the magnetoresistance, the coherence temperature may be slightly smaller than T_K. It may be noted that there is a relatively steeper fall in the value of inverse χ for $x=0.53$ compared to $x=0.63$ and the value for the latter is about 3 times larger at 5 K. Since the low-temperature electronic terms in C are not widely different for the two alloys, we believe that the low-temperature enhancement of χ for $x=0.53$ arises from some traces of magnetic impurities, which may be consistent with a relatively larger intensity of the 6K peak in Fig. 4 for this composition.

Summarizing, we have presented to ρ, $\Delta\rho$, C and χ data for the alloys, $CeFe_xGe_2$. We have critically looked at the origin of $T_{\max}$ and it appears that a crossover from an incoherent to coherent region (as the temperature is lowered) around $T_{\max}$ is highly probable in these alloys, though these alloys are characterized by a number of vacant interstitial sites. If this is true, the coherence is maintained due to the ordered replacement of Ni sites (in $CeNi_xSi_2$ structure) by Fe. We believe that these systems would contribute to the generalization of Kondo-lattice behavior in disordered compounds.

Two of us knowledge the support of R. Vijayaraghavan.

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