Thermodynamic and transport properties of $(Ce_xGd_{1-x})Cu_6$ for $0 \leq x \leq 1$

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Results are reported for the properties of the pseudobinary alloys between the heavy-fermion compound $CeCu_6$ and the frustrated antiferromagnet $GdCu_6$. Evidence is provided for a rapid onset of Kondo behavior in the resistivity for small Ce substitutions in $GdCu_6$, the persistence of signs of Kondo coherence for significant substitutions of Gd in $CeCu_6$, and evolution through some type of spin-glass or spin-density-wave magnetic freezing to local-moment magnetism as the Gd content increases from 10% to 90%. An interesting possibility exists that in the Ce-rich concentration regime the freezing is the stabilization of some type of spin-density wave in the Ce magnetization, as produced by Au on the Cu sites or in the yttrium conduction band by Gd substitutions in pure Y. As in the resistivity, Gd moment contributions and Kondo (Ce) contributions seem to be independent and additive in the entropy.

I. INTRODUCTION

The importance of magnetic correlations in heavy-fermion compounds is now well recognized.[1] Short-range correlations, in fact, are expected to be present even if the paramagnetic Fermi liquid (which heavy-fermion compounds can be described as) remains stable at low temperatures[2] and are necessary for quenching of local moments in the lattice,[3] a manifestation of which is the Kondo-like behavior observed in the heavy fermion compounds. But it is becoming more and more clear that apart from the heavy-fermion compounds which actually order antiferromagnetically, e.g., UCd_{11} (Ref. 4) and U_2Zn_{17},[5] longer-range magnetic correlations in some form or other are present in many other heavy-fermion systems. For example, in UPt_3 evidence now exists for some sort of antiferromagnetic order around 6 K, and more clearly defined antiferromagnetism readily establishes itself with very small doping by Th on U sites[6] and Pd on Pt sites.[7] A similar type of weak magnetic ordering is believed to be present in $CeAl_3$ (Ref. 8) and $CeCu_2Si_2$ also.[9] In UBe_{13} the occurrence of a double transition with Th doping is a very well-studied phenomenon[10] and there exists evidence that the lower transition can be associated with a spin-density-wave-(SDW-) type magnetic ordering,[11] while the upper one is a superconducting transition. Very recently, magnetostriction measurements have suggested antiferromagnetic ordering at 8.8 K in UBe_{13} itself.[12]

In the heavy-fermion system $CeCu_6$, although no long-range magnetic ordering has so far been observed down to 20 mk of temperature, its nearness to magnetic behavior is evident in the effects of doping with Ag,[13] Au,[14] and Pt.[15] It should be mentioned, however, that antiferromagnetic spin fluctuations of short-range nature are present in undoped $CeCu_6$ at very low temperature ($T=0.4$ K), as evidenced from neutron measurements.[16] On the other hand, it has now been observed from resistivity and specific-heat measurements that in isostructural RCu_6 compounds, where $R=$La (Ref. 17) and Pr,[18] a small substitution of Ce readily gives rise to Kondo behavior. It is our interest here, starting with the isostructural compound $GdCu_6$ which (unlike the previously studied RCu_6 compounds) orders antiferromagnetically at $T=16$ K, and gradually substituting Gd with Ce to study the onset of Kondo behavior and its interaction with magnetism as one approaches the $CeCu_6$ end. The magnetic properties of the end compound $GdCu_6$ are also quite interesting. Although we shall mention that (where relevant), in connection with our present work, more emphasis actually will be placed here on the Kondo behavior, keeping in mind the eventual emergence of a heavy-fermion ground state in $CeCu_6$.

In the subsequent sections we present the results of our resistivity, ac susceptibility, dc magnetization, and specific-heat measurements on $(Gd_{1-x}Ce_x)Cu_6$ and seek an interpretation within the realm of existing theoretical models.

II. EXPERIMENT

The alloys were prepared by argon arc melting and subsequent suction chill casting into copper moulds to

produce square cross-section rods. The samples were subjected to metallographic analysis to investigate the possible presence of a second phase. A very small amount of second phase (certainly less than 5% and not detectable by x ray) is observed in some of the samples, but there is no evidence that this has any significant effect on the properties we have studied. An additional check for the phase purity of the samples is provided by our small field (0.7 G) ac susceptibility measurements. The second-phase compounds (as distinct from a copper-based solid solution) most likely to be present in our samples are $CeCu_5$ and $GdCu_5$. Both $GdCu_5$ and $CeCu_5$ order antiferromagnetically at 25 and 3.5 K, respectively, but with our ac susceptibility apparatus, which in past has revealed small magnetic impurities,[19,20] we could not detect any signature attributable either to $GdCu_5$ or to $CeCu_5$. Some of the samples in the intermediate concentration regime ($x=0.9$, 0.75, 0.5, and 0.25) were subjected to x-ray-diffraction analysis to check whether they have the same structure as the parent compounds. All of them show diffraction lines associated with the orthorhombic structure of the parent compounds.

The resistivity of the samples of typical dimension ($10m \times 1.5m \times 1.5mm^3$) was measured using a standard four-probe method, ac susceptibility measurements were performed with a driving frequency of 320 Hz and a driving field of 0.7 G. A quantum design superconducting quantum interference device magnetometer was used to measure dc magnetization and susceptibility. The specific heat of the samples between 1.1 and 18 K was measured using a time constant method.[21,22]

III. RESULTS

A. Resistivity

Resistivity (ρ) results for $(Ce_xGd_{1-x})Cu_6$ with $x=0$, 0.1, 0.25, 0.5, 0.9, and 1 are shown in Figs. 1 and 2. Results exist for $x=0.4$, 0.6, 0.75, and 0.95, but are not shown here for the sake of clarity. Error bars for the data are 10%, primarily due to uncertainties in the cross-sectional area of the bar and in the voltage contact separation. Results for the end compounds, i.e., $CeCu_6$ and $GdCu_6$, agree well with the existing results.[23,24] The striking feature of our results is the appearance of a Kondo-type minimum in the ρ-versus-T plot with a mere 10% substitution of Ce in $GdCu_6$, followed by a sharp knee at a lower temperature. The latter feature is the characteristic of freezing of spin disorder scattering associated with a magnetic transition. This lower-temperature magnetic transition (which occurs at a temperature slightly lower than that of $GdCu_6$) is confirmed by our subsequent measurements of ac and dc susceptibility. To check whether the resistivity minimum is an effect of Ce substitution or in some way related to the magnetic ordering in the system (although no such minimum was observed as a precursor of magnetic ordering in $GdCu_6$), we measured the resistivity of $(Gd_{0.9}Y_{0.1})Cu_6$ and $(Gd_{0.5}Y_{0.5})Cu_6$ (shown in Fig. 3), which show only a knee in the ρ-versus-T curve without any hint of a minimum. This suggests that the Ce ions, at least at higher temperatures, act as independent scattering centers and give rise to Kondo-like behavior. With further increase in Ce concentration, the resistivity minimum gains in strength very rapidly and the temperature of the minimum (T_{min}) is elevated, while the low-temperature knee becomes much weaker and the corresponding transition temperature decreases. With an increase in x, the sharp knee in $GdCu_6$ becomes just a change in slope for $x=0.25$ and that too is almost erased by $x=0.5$. With further increase in Ce concentration, no structure (i.e., knee or change in slope) in the ρ-versus-T plots is visible below T_{min} until $x=0.9$, where a rounded resistivity maximum appears at low temperature (T_{max}). This becomes more prominent, and T_{max} increases as one approaches the $CeCu_6$ end. This maximum (which is a characteristic of almost all heavy-fermion compounds) is often termed the Kondo coherence peak, and it is suggested to be a precursor of coherence between the $4f$ (Ce) scattering sites at low temperature, the logarithmic behavior at higher temperature being taken as the hallmark of an independent-ion Kondo effect (see Ref. 1). It is interesting to note that all these heavy-fermion features of

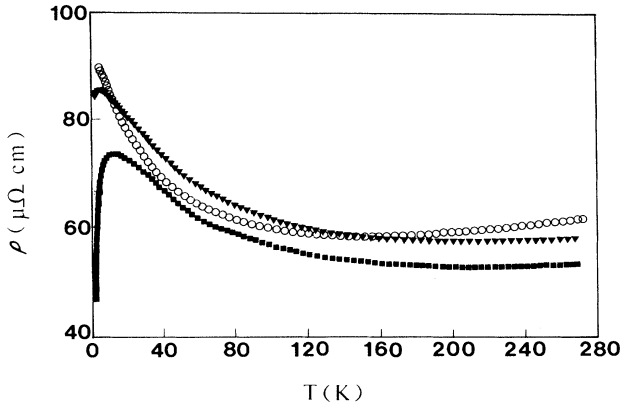

FIG. 1. Resistivity (ρ) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ■, $x=1$; ▼, $x=0.9$; ○, $x=0.5$.

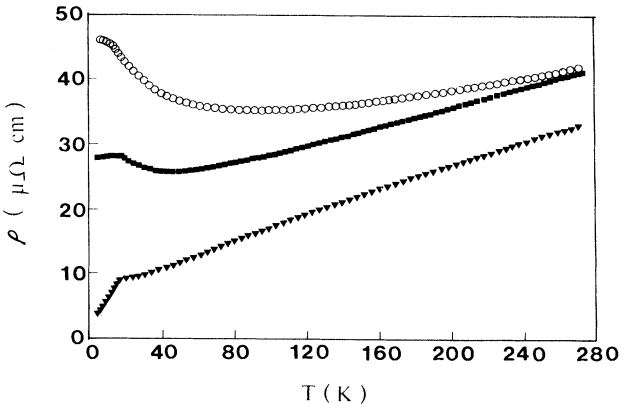

FIG. 2. Resistivity (ρ) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ○, $x=0.25$; ■, $x=0.1$; ▼, $x=0$.

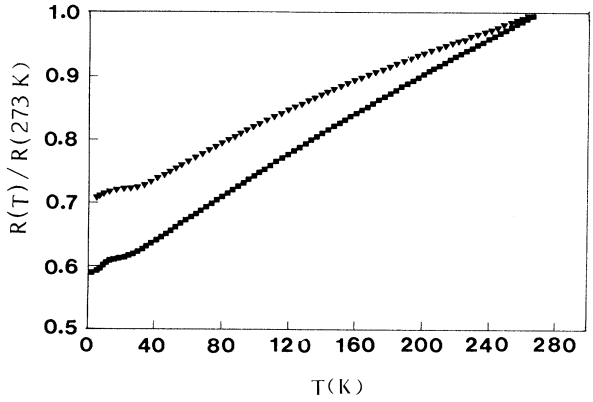

FIG. 3. Normalized resistivity $R(T)/R(273)$ as a function of temperature (T) for $(Y_xGd_{1-x})Cu_6$: ■, $x=0.1$; ▼, $x=0.5$.

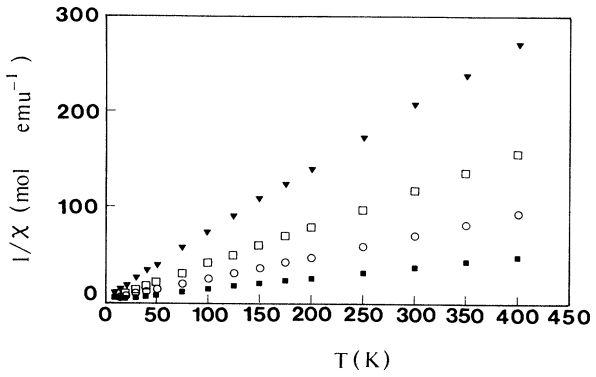

FIG. 4. Reciprocal dc susceptibility ($1/\chi$) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ■, $x=0$; ○, $x=0.5$; □, $x=0.75$; ▼, $x=0.9$.

resistivity in $CeCu_6$ are retained up to 10% Gd substitution. Our present results, along with those on $(Ce,T)Cu_6$, where T=La, and Pr (Refs. 17 and 18) and Ce $(Cu,T)_6$, where T=Au, Ag, Al, and Pt,[13–15] establish a very distinct trend in the effects of alloying in $CeCu_6$. A substitution of at least 10% at the Ce site is required to erase the Kondo coherence peak, and the single or independent-ion Kondo behavior is present quite prominently even with 90% substitutions. This latter effect is particularly interesting in the case of Gd substitutions, where in the Gd-rich regime there are clear indications of magnetic ordering at low temperature. On the other hand, 2–3 % substitutions at the copper site by nonmagnetic elements such as Au, Ag, Al, and Pt are sufficient to erase the Kondo coherence peak, and further substitutions (5–10 %) cause strong suppression even of the single-ion Kondo behavior.

B. dc susceptibility

Figure 4 shows the inverse susceptibility ($1/\chi$) versus temperature (T) plots of $(Ce_xGd_{1-x})Cu_6$ with $x=0$, 0.5, 0.75, and 0.9, measured in a field of 5 kG. Results also exist for $x=0.1$, 0.6, and 1, but are not shown here for the sake of clarity. A Curie-Weiss law is obeyed in all of the samples in the temperature regime above 100 K. Effective moment values of the samples (μ_{eff}) calculated from the linear fit of the ($1/\chi$) plots above 150 K are given in Table I. The values of μ_{eff} for the end compounds $CeCu_6$ and $GdCu_6$ are $2.63\mu_B$ and $8.1\mu_B$, respectively, which are within 10% of the literature values. In the intermediate-concentration regime, there is a gradual change in μ_{eff} between these two numbers, which seems to support the indication from the resistivity results that at high temperatures Ce and Gd ions contribute independently to the various physical processes. In Fig. 5 we plot the susceptibility (χ) results between 2 and 25 K. $GdCu_6$ displays its antiferromagnetic peak at 16.5 K, which is in accord with earlier results. With 10% Ce substitution, i.e., $x=0.1$, the nature of the peak essentially remains the same, but the magnetic ordering temperature (T_{ord}) decreases slightly. The ordering temperature of this alloy, along with the others in the series, is given in Table I. T_{ord} for $(Ce_{0.5}Gd_{0.5})Cu_6$ is around 8 K, but there is a marked change in the character of the susceptibility. Instead of showing a typical antiferromagnetic

TABLE I. Effective moments and ordering temperatures, obtained from dc susceptibility (χ_{dc}), ac susceptibility (χ_{ac}), resistivity (ρ) and specific-heat (C_p) measurements for $(Ce_xGd_{1-x})Cu_6$.

Compounds	μ_{eff} (μ_B per formula unit)	T_{ord} (K) (dc χ)	T_{ord} (K) (ac χ)	T_{ord} (K) (ρ)	T_{ord} (K) (C_p)
$GdCu_6$	8.12	16.8	16.4	17.2	15.8[a]
$x=0.1$	7.98	15	14.9	15.5	
$x=0.25$			12.5	13.5	11.9
$x=0.5$	5.973	8.7	9.7	9.8	8.3
$x=0.6$		6.2	7.5	7.8	6.8
$x=0.75$	4.566	3.2	3.6		4.1
$x=0.9$	3.5				1.8
$CeCu_6$	2.63				

[a]From Ref. 24.

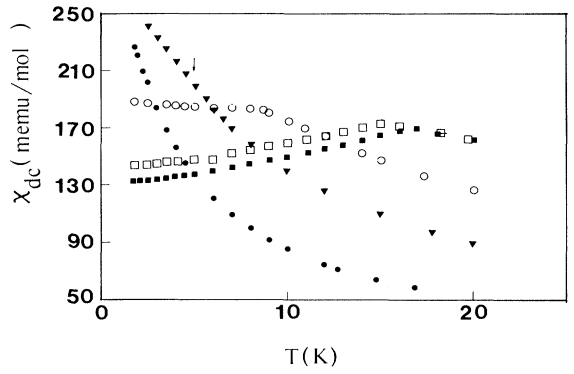

FIG. 5. dc susceptibility (χ) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ■, $x=0$; □, $x=0.1$; ○, $x=0.5$; ▼, $x=0.75$; ●, $x=0.9$.

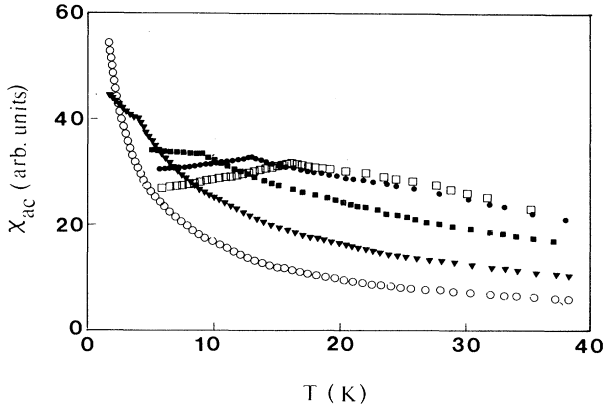

FIG. 7. ac susceptibility (χ_{ac}) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: □, $x=0$; ●, $x=0.25$; ■, $x=0.5$; ▼, $x=0.75$; ○, $x=0.9$.

peak, the susceptibility remains almost flat below T_{ord}, and with further increase in Ce concentration the only indication of magnetic order or freezing is a distinct change in slope in the χ-versus-T plots; for $x=0.75$ even this change in slope is barely visible (its position is indicated by the arrow in Fig. 5). These results suggest that the nature of magnetic ordering in the alloys with $x \geq 0.5$ is different from that in the Gd-rich regime. To check the possibility of a spin-glass-type magnetic ordering, we have measured the zero-field-cooled (ZFC) and field-cooled (FC) susceptibility of the $x=0.6$ alloy in a field of 50 G (see Fig. 6). Although there is a difference in ZFC and FC behavior below T_{ord}, this is not as prominent as is observed in typical spin glasses.

C. ac susceptibility

We present in Fig. 7 our low-field (0.7-G) ac susceptibility results for $(Ce_xGd_{1-x})Cu_6$ with $x=0$, 0.25, 0.5, 0.75, and 0.9 between 2 and 40 K. Results exist for $x=0.4$ and 0.6, but are not shown for the sake of clarity. The susceptibility behavior for $x \leq 0.6$ is quite similar to that obtained from much higher-field (5000-G) dc susceptibility, thus ruling out any straightforward spin-glass-like behavior in the alloys with $x=0.5$ and 0.6. In the alloy with $x=0.75$, a sharp kink is now visible, which was quite smeared out in the higher-field dc susceptibility measurements. No distinct sign of any magnetic transition is visible for $x=0.9$ down to 2 K.

From our dc and ac susceptibility measurements, it appears that the nature of the magnetic ordering of the alloys with $x>0.5$ is different from the antiferromagnetic ordering observed in the Gd-rich regime, but this difference is subtle and one needs measurements of frequency-dependent and nonlinear susceptibility and various microscopic measurements to clarify the situation.

D. dc magnetization

Results of our magnetization measurements in fields up to 5.5 T (at 2 K) for the alloys with $0<x<1$ are shown in Figs. 8 and 9. In $GdCu_6$ there is a distinct upward curvature around $H=2.5$ T, indicating the presence of a spin-

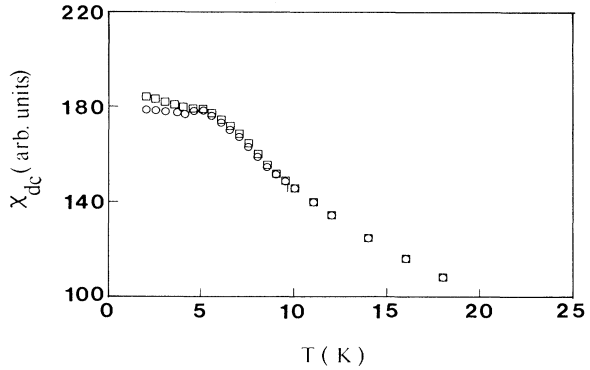

FIG. 6. dc susceptibility (χ) as a function of temperature (T) for $(Ce_{0.6}Gd_{0.4})Cu_6$: ○ = zero-field cooled; □ = field cooled.

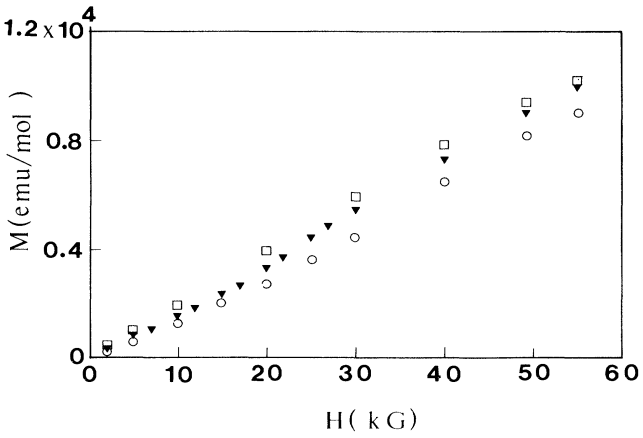

FIG. 8. Magnetization (M) at 2 K as a function of magnetic field (H) for $(Ce_xGd_{1-x})Cu_6$: ○, $x=0$; ▼, $x=0.1$; □, $x=0.5$.

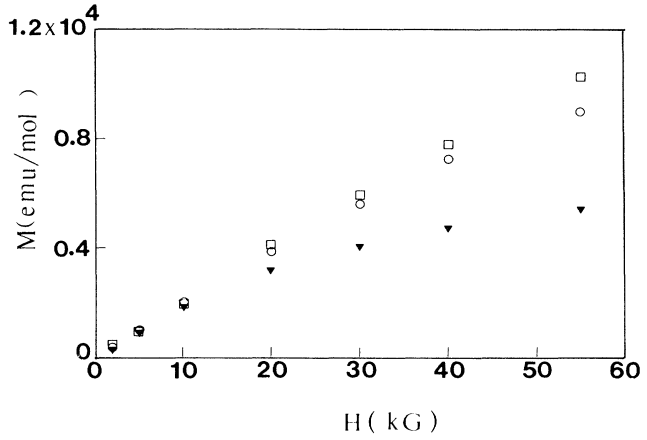

FIG. 9. Magnetization (M) at 2 K as a function of magnetic field (H) for $(Ce_xGd_{1-x})Cu_6$: □, $x=0.5$; ○, $x=0.75$; ▼, $x=0.9$.

flip-type or metamagnetic transition. This interesting aspect of $GdCu_6$ was earlier pointed out by Takayanagi *et al.*[24] in their study of single crystals of $GdCu_6$. In our $x=0.1$ sample, the metamagnetic transition seems to occur at a lower field and there is only a weak indication of such a transition in $x=0.5$. It is to be noted that the values of magnetization for $x=0.1$ and 0.5 are somewhat higher than that of $GdCu_6$. On quantitative consideration of our dc and ac susceptibility results, we find that the magnitude of susceptibility at and below the ordering temperature is higher in $x=0.1$ and 0.5 than in $GdCu_6$. There is a slight nonlinearity in the magnetization (M) versus field (H) curve for $x=0.5$ which becomes more visible for $x=0.75$ and 0.9. In another system $[(Ce,Gd)Al_3]$,[25] similar differences in the nature of the M-versus-H curve between Gd- and Ce-rich regions has been observed where it was thought to be indicative of the different nature of magnetic ordering in the two regimes.

E. Specific heat

Specific-heat results for $(Ce_xGd_{1-x})Cu_6$ alloys with $x=0.25$, 0.5, 0.6, 0.75, and 0.9 are shown in Fig. 10. A peak indicative of some sort of magnetic ordering is visible in all of the samples. The height of the peak and the associated ordering temperatures decrease with increase in Ce concentration, the latter effect being in accord with our resistivity and magnetic measurements. We wish to emphasize the difference in the nature of magnetic ordering between the Ce- and Gd-rich concentration regime. The specific-heat behavior of the alloy $x=0.25$ retains essentially all the characteristic of the end compound $GdCu_6$,[24] namely, a λ-type anomaly at the transition temperature followed by broad hump at a lower temperature. This broad hump, which cannot be explained simply in terms of crystal-field effects, indicates the complexity of the magnetic excitations below the ordering temperature. An analogy can be drawn here with GdB_6 which also shows a λ-type anomaly around 18 K followed by another transition at 6 K, the origin of which is not very clear at the moment.[26] Another interesting aspect of $GdCu_6$ is the fact that the specific-heat anomaly is not of a simple λ type, but consists of two distinct peaks at 15.8 and 16.3 K.[24] Similar multiple-peak structures have also been observed in other isostructural compounds, namely, $NdCu_6$ and $SmCu_6$.[27] Within the experimental uncertainties there seems to be the presence of a similar structure in the specific-heat peak of $x=0.25$. It should be noted, however, that no indication of a double magnetic transition was found in the resistivity and magnetic susceptibility studies, either in this sample or in pure $GdCu_6$.

The specific-heat result for $x=0.75$ is very different from that of $x=0.25$. The maximum is much broader and has a long tail above the peak temperature. Such a behavior might arise from sample inhomogeneity, but such an origin is not very likely since this sample was annealed at 700 °C for a week. In conjunction with specific-heat results of the other alloys along with the resistivity and magnetic susceptibility results, one can infer that the observed specific-heat behavior in $x=0.75$ is quite similar to that observed in various rare-earth-based spin glasses, although a similarity to local-

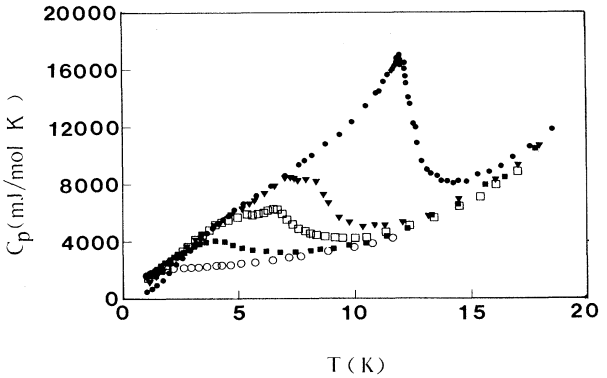

FIG. 10. Specific heat (C_p) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ●, $x=0.25$; ▼, $x=0.5$; □, $x=0.6$; ■, $x=0.75$; ○, $x=0.9$.

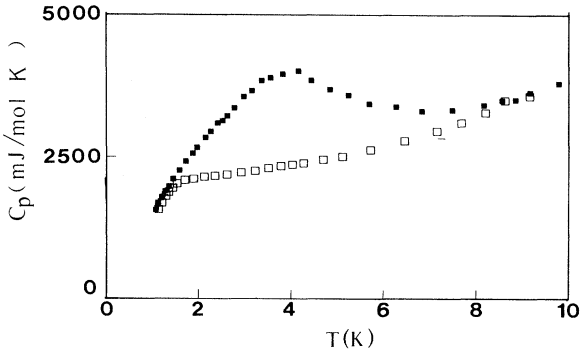

FIG. 11. Specific heat (C_p) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$ with $x=0.75$ (■) and $x=0.9$ (□). This is to highlight the differences between these two compounds.

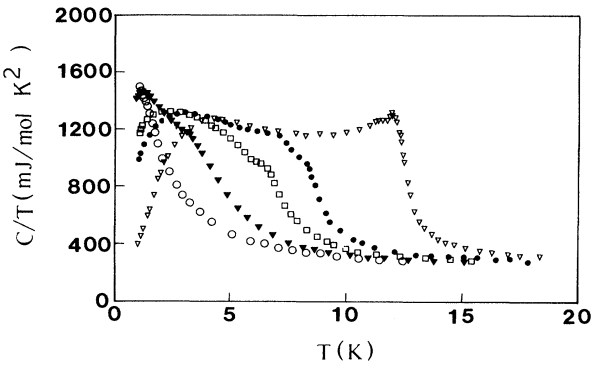

FIG. 12. Specific heat divided by temperature (c_p/T) as a function of temperature (T) for $(Ce_xGd_{1-x})Cu_6$: ▽, $x=0.25$; ●, $x=0.5$; □, $x=0.6$; ▼, $x=0.75$; ○, $x=0.9$.

moment-stabilized spin-density-wave systems such as $Y_{0.978}Gd_{0.022}$ (Ref. 28) cannot be ruled out. In this latter system it is now concluded from a combination of neutron-scattering, calorimetry, and susceptibility measurements that the Gd local moments undergo a coupling via a long-range helical spin-density-wave ordering of the band electrons. In Fig. 11 we compare the specific-heat results of $x=0.75$ and 0.9. The structure in the specific heat for $x=0.9$ around 1.5 K is quite similar to that observed in UBe_{13} around 3 K,[29] which may be connected with the suggested magnetic ordering, although the latter is around 8 K.[12] In conjunction with the results on other alloys, in the present system it seems likely that the $x=0.9$ behavior is due to magnetic ordering of spin-glass type. However, it should be mentioned here that in another system $[Ce(Al_{1-x}Cu_x)_2]$, while specific-heat results showed a broad hump, there was no indication of any magnetic ordering in the low-field ac susceptibility data in the same temperature regime.[30] Having identified a difference in the nature of magnetic ordering between the Ce- and Gd-rich regions, from Fig. 10 we can regard the magnetic character of the $x=0.5$ and 0.6 alloys as intermediate between that of those with $x=0.25$ and 0.75.

We plot in Fig. 12 C/T-versus-T plots for $x=0.25$, 0.5, 0.6, 0.75, and 0.9. In $x=0.25$ the sharp drop in C/T at low temperature seems to be associated with the specific-heat hump in the same temperature region. With an increase in Ce concentration, the anomaly in C/T around T_N became progressively less sharp and hardly visible in the case of $x=0.75$, while the temperature at which the low-temperature drop occurs was pushed toward lower temperature. In the case of $x=0.9$, the behavior of C/T is rather smooth down to 1.1 K. It is to be noted that the temperature dependence of C/T in the alloys described above is quite different from that of the other well-studied heavy-fermion antiferromagnets, i.e., U_2Zn_{17} and UCd_{11}. On a closer look, however, there appears to be a hint of a shoulder below T_N of UCd_{11},[29] and a recent pressure study suggests the possibility of more magnetic transitions in the low-temperature regions.[31]

In Table II we present our estimated magnetic entropy associated with magnetic transitions in $(Ce_xGd_{1-x})Cu_6$. We have subtracted the specific heat of YCu_6 as nonmagnetic background and calculated the entropy from the area under the C/T-versus-T plots with the upper limit of T as $1.2T_{ord}$. Since $CeCu_6$ does not order magnetically (down to the lowest attainable temperature), we have estimated its entropy by choosing an upper limit of T (somewhat arbitrarily) as 6 K. Entropy associated with the antiferromagnetic structure of $GdCu_6$ has been taken from Ref. 24. This is found to be 15 J/mol K and in accord with the expected ground state of $^8S_{7/2}$ for the Gd ion. From Table II we find that the estimated entropy S (=5443 mJ/mol K) for the alloy $x=0.75$ is higher than that expected from the contribution (3850 mJ/mol K) of Gd ions alone. On the other hand, if the entropy consists of contributions from both Ce and Gd (in appropriate proportions), then the estimated value of entropy comes within 10% of the expected value. In the alloy with $x=0.25$, although Gd alone is still unable to account for all the estimated entropy, 90% of the entropy can now be attributed to them. This picture is in accord with our resistivity study where we have seen a progressive increase in Kondo behavior with the increase in Ce concentration.

IV. DISCUSSION

The present work can be regarded as related to previous work on the systems such as (U,Gd)Al_2 (Ref. 32) and

TABLE II. Estimated magnetic entropy associated with the magnetic transitions in $(Ce_xGd_{1-x})Cu_6$.

Compound	S_1 (mJ/mol K)[a]	S_2 (mJ/mol K)[b]	S_3 (mJ/mol K)[c]
$CeCu_6$	2760	2760	5763
$x=0.9$	3555	4024	6916
$x=0.75$	5443	5920	8644
$x=0.6$	7547	7816	10 373
$x=0.5$	9054	9080	11 526
$x=0.25$	12 724	12 990	14 408

[a] S_1 = entropy estimated from experimental data, i.e., Fig. 12.
[b] $S_2=xS(CeCu_6)+(1-x)S(GdCu_6)$, where $S(CeCu_6)=2760$ (from present measurement) and $S(GdCu_6)=15\,400$ (from Ref. 24).
[c] $S_3=xR\ln 2+(1-x)R\ln 8$; this is the theoretically estimated entropy, where $R\ln 2$ is the entropy associated with doublet ground state of $CeCu_6$ and $R\ln 8$ is associated with the expected ground state of $^8S_{7/2}$ in the Gd^{3+} ion of $GdCu_6$.

$(Ce,Gd)Al_3$ (Ref. 33) where one crosses continuously from an RX_n heavy-fermion system to a magnetically ordered GdX_n system, ferromagnetically in $GdAl_2$ and antiferromagnetically but strongly frustrated in $GdAl_3$.[34] However, UAl_2 is only very modestly a heavy-fermion system and the metallurgical problems created by the peritectoid character of $CeAl_3$ make for some difficulties of interpretation in $(Ce,Gd)Al_3$.[33]

The main questions that arise in the present work are as follows.

(a) How far does the substitution of moment-bearing Gd atoms on the Ce sites affect the heavy-fermion character of $CeCu_6$?

(b) Are the specific-heat contributions of the Ce heavy fermions and those of the Gd moments simply additive?

(c) Does the magnetic freezing for smaller Gd concentrations have the simple spin-glass character of $(U,Gd)Al_2$ or does the alloying on the Ce sites facilitate some type of spin-density-wave order of the Ce $4f$ hybrid states in the way that happens for Ag or Au substitutions on the Cu sites?[13,14]

(d) Is the independent or single-ion Kondo behavior that seems to be known for smaller Ce concentrations similar to that for $(La,Ce)Cu_6$ (Ref. 17) and insensitive to the Ruderman-Kittle-Kasuya-Yosida (RKKY) mediated coupling of Gd moments?

Definitive answers to all these questions cannot yet be given, but a few pointers exist.

(a) As with substitutions on the Cu sites, suppression of coherence in the resistivity is fairly clear; it seems to take place more rapidly than for nonmagnetic substitutions on the Ce site, but less rapidly than for substitutions on the Cu sites.

(b) Even at 10% Gd, there seems to be a signature of magnetic freezing effects at low temperature in the specific heat, but there are indications that a heavy-fermion contribution coexists with these. At the Gd-rich end, calculations of the $4f$ magnetic entropy suggest that a single-ion Kondo contribution is made by Ce. These remarks are supported by the data shown in Table II.

(c) The temperatures of magnetic freezing indicated by our measurements suggest that these do not diverge greatly from those that would be given by simple dilution of Gd-Gd RKKY interactions. However, the specific-heat behavior in the Ce-rich region does not correspond exactly to that found in canonical spin glasses like *Cu*Mn and seems to have something in common at least at 25% Gd with the behavior of dilute *Y*Gd alloys where a long-range incommensurate spin-density wave is stabilized by the local moments.[28] One is encouraged to take this interpretation more seriously by the recent observation[35] that the magnetic order stabilized by small amount of Au substitutions is incommensurate. It is interesting to note that in our $(Ce_{0.5}Gd_{0.5})Cu_6$ sample the plot of C_M/T as a function of T is very similar in character to that of Y 4.4% Gd.[28]

(d) The observation of a resistivity minimum in the whole composition range suggests the presence throughout of incoherent Kondo scattering, but the temperature of the minimum decreases steadily from about 220 K in pure $CeCu_6$ to ≈ 90 K at $(Ce_{0.25}Gd_{0.75})Cu_6$ and ≈ 50 K at $(Ce_{0.1}Gd_{0.9})Cu_6$ in which it is found above the knee associated with antiferromagnetic ordering.

We believe we have shown that the magnetic behavior of $(Ce_xGd_{1-x})Cu_6$ has quite interesting features, in particular the presence of strong Kondo behavior across the whole concentration regime. Further experiments, especially microscopic ones, are required to ascertain the exact nature of various magnetic orderings.

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[1]N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1990), Vol. 14.

[2]M. R. Norman, T. Oguchi, and A. J. Freeman, Phys. Rev. B **38**, 11 193 (1988); also various references quoted in Ref. 1.

[3]P. Nozieres, Ann. Phys. (Paris) **10**, 19 (1985).

[4]Z. Fisk, G. R. Srewart, J. O. Willis, H. R. Ott, and F. Hulliger, Phys. Rev. B **30**, 6360 (1984).

[5]H. R. Ott, H. Rudigier, P. Delsing, and Z. Fisk, Phys. Rev. Lett. **52**, 1551 (1984).

[6]G. R. Stewart, A. L. Giorgi, J. O. Willis, and J. O. O'Rourke, Phys. Rev. B **24**, 4269 (1986).

[7]A. de Visser, J. C. P. Klasse, M. van Spang, J. J. M. Franse, A. Menovsky, and T. T. M. Palstra, J. Magn. Magn. Mater. **56-57**, 375 (1986).

[8]S. Barth, H. R. Ott, F. W. Gygax, B. Hitti, E. Lippelt, A. Schenck, and C. Baines, Phys. Rev. B **39**, 11 695 (1989).

[9]H. Nakamura, Y. Kitaoka, H. Yamada, and K. Asayama, J. Magn. Magn. Mater. **76-77**, 517 (1988).

[10]H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. B **31**, 1651 (1985).

[11]B. Batlogg, D. B. Bishop, B. Goldring, C. M. Varma, Z. Fisk, J. L. Smith, and H. R. Ott, Phys. Rev. Lett. **55**, 1319 (1985); R. H. Heffner, S. W. Cooke, and D. E. MacLaughlin, in *Theoretical and Experimental Aspects of Valence Fluctuations and Heavy Fermions,* edited by L. C. Gupta and S. K. Malik (Plenum, New York, 1987), p. 319.

[12]R. N. Kleiman, D. J. Bishop, H. R. Ott, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **64**, 1975 (1990).

[13]A. K. Gangopadhyay, J. S. Schilling, E. Schuberth, P. Gutsmeidt, F. Gross, and K. Andreas, Phys. Rev. B **38**, 2603 (1988); G. Fraunberger, B. Andraka, J. S. Kim, U. Ahlheim, and G. R. Stewart, *ibid.* **40**, 4735 (1989).

[14]A. Germann, A. K. Nigam, J. Dutzi, A. Schroder, and H. v. Lohneysen, J. Phys. (Paris) Colloq. **49**, C8-755 (1988); M. R. Lees, B. R. Coles, E. Bauer, and N. Pillmayer, J. Phys: Condens. Matter **2**, 6403 (1990).

[15]S. B. Roy, G. R. Stewart, and B. R. Coles (unpublished).
[16]G. Aeppli, H. Yoshizawa, Y. Endoh, and T. Komatsubara, Phys. Rev. Lett. **57**, 122 (1986).
[17]K. Satoh, T. Fujita, Y. Maeno, Y. Oniki, and T. Komatsubara, J. Phys. Soc. Jpn. **58**, 1012 (1989).
[18]S. B. Roy, G. R. Stewart, and B. R. Coles, J. Magn. Magn. Mater. (to be published).
[19]S. B. Roy, J. Phys.: Condens. Matter **1**, L165 (1989).
[20]J. A. Chilton, Ph.D. thesis, University of London, 1988.
[21]R. Bachmann, F. J. Di Salvo, T. H. Geballe, R. L. Greene, R. E. Howard, C. N. King, H. C. Kirsch, K. N. Lee, R. E. Schwall, H. U. Thomas, and R. B. Zubeck, Rev. Sci. Instrum. **43**, 205 (1972).
[22]G. R. Stewart, Rev. Sci. Instrum. **54**, 1 (1983).
[23]H. R. Ott, H. Rudigier, Z. Fisk, J. O. Willis, and G. R. Stewart, Solid State Commun. **53**, 235 (1985).
[24]S. Takayanagi, Y. Onuki, K. Ina, T. Komatsubara, N. Wada, T. Watanabe, T. Sakakibara, and T. Goto, J. Phys. Soc. Jpn. **58**, 1031 (1989).
[25]A. S. Edelstein and R. L. Holtz, J. Appl. Phys. **63**, 3689 (1988).
[26]S. Kunii, K. Takeuchi, I. Oguro, K. Sugiyama, A. Ohya, M. Yamada, Y. Koyoshi, M. Date, and T. Kasuya, J. Magn. Magn. Mater. **52**, 275 (1985).
[27]S. Takayanagi, Y. Onuki, T. Komatsubara, N. Wada, and T. Watanabe, Jpn. J. Appl. Phys. **26**, 535 (1987).
[28]L. E. Wenger, G. W. Hunter, A. Mydosh, J. A. Gotaas, and J. J. Rhyne, Phys. Rev. Lett. **56**, 1090 (1986).
[29]G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).
[30]M. R. Lees and E. Bauer (unpublished).
[31]J. D. Thompson, Z. Fisk, M. W. McElfresh, H. R. Ott, and M. B. Maple, Phys. Rev. B **39**, 2568 (1989).
[32]J. Y. Ping and B. R. Coles, J. Magn. Magn. Mater. **29**, 209 (1982); B. R. Coles, J. Y. Ping, and M. H. Bennett, Philos. Mag. B **50**, 1 (1984).
[33]A. S. Edelstein, R. L. Holtz, D. J. Gillespie, M. Rubenstein, J. J. Tyson, R. A. Fisher, and N. E. Phillips, Phys. Rev. B **37**, 7877 (1988).
[34]B. R. Coles, S. Oseroff, and Z. Fisk, J. Phys. F **17**, L169 (1987).
[35]T. Chattopadhyay, H. v. Lohneysen, T. Trappman, and M. Lowenhaupt, Z. Phys. B **80**, 159 (1990).