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# Anomalous low-temperature properties of dilute Ce alloys $Ce_xLa_{1-x}Cu_{2.2}Si_2$ $(x \leq 0.2)$

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Specific heat, magnetic susceptibility, and electrical resistivity have been investigated for moderately dilute $Ce_xLa_{1-x}Cu_{2.2}Si_2$ alloys $(x \leq 0.2)$ between 1 and 20 K. $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ has been found to possess a logarithmic temperature variation in $C/T$ and $\chi$, implying a possibility of the two-channel Kondo effect. However, magnetic-field dependence of $C$ is not consistent with such an interpretation. Anomalous spin-glass-like behavior observed for related $Ce_{0.15}La_{0.85}Cu_{2.2}Si_2$ at low temperatures indicates a magnetic nature of the phenomenon.

Recently, several U-based alloys[1-6] have been found to obey a logarithmic or power-law temperature dependence of their low-temperature specific heats divided by temperature. Such a behavior is clearly inconsistent with theories based on a Fermi-liquid hypothesis, one of the most fundamental canons in condensed-matter physics. This observation revived also more than a decade-old question about an origin of the low-temperature enhancement in thermodynamic and magnetic properties of materials, somewhat loosely classified as heavy fermions.[7] An outstanding majority of heavy-fermion systems are based on one of the two elements, Ce or U. It has been fairly well accepted that the origin of this enhancement in Ce alloys derives from a Kondo effect.[8] The Kondo model, which is a single-impurity model, has a Fermi-liquid ground state.

$CeCu_2Si_2$ (or $CeCu_{2.2}Si_2$; superstoichiometric Cu stabilizes the superconducting state below 0.6 K) is usually considered an archetypal Kondo-lattice system,[9] i.e., a system in which interactions between different Kondo centers result in some deviations from the single-impurity behavior. These intersite interactions should gradually diminish when concentration of the Kondo centers decreases. Therefore, we have investigated the low-temperature properties of $Ce_xLa_{1-x}Cu_{2.2}Si_2$ for somewhat dilute ranges of concentrations, $x=0.2$, 0.15, 0.1, and 0.05. Investigation of more dilute alloys was not attempted, for large uncertainties in the $f$-electron specific heat, magnetic susceptibility, and electrical resistivity derived for such alloys. This work is an extension of a more systematic study already reported.[10]

The $f$-electron specific heat for each alloy was obtained by subtracting the total specific heat of $LaCu_{2.2}Si_2$. No corresponding subtraction was performed for the magnetic susceptibility of the investigated alloys since $LaCu_{2.2}Si_2$ has very small diamagnetic susceptibility which is essentially temperature independent. Throughout this paper, $C$ and $\chi$ stand, respectively, for the $f$-electron specific heat and magnetic susceptibility, both normalized to a mole of Ce.

The overall trends in the specific heat upon change of the Ce concentration $x$, for $x \rightarrow 0$, can be inferred from Fig. 1. $C/T$ versus $\log_{10}T$ is shown for $x=0.025$, 0.1, 0.15, and for temperatures between 1 and 10 K. In agreement with the results of Ref. 10 $C/T$ values at $T=1.1$ K increase with a decrease of $x$ for $x<0.5$. We have found the largest rate of increase between $x=0.2$ and $x=0.1$, followed by a tendency toward saturation for the concentrations corresponding to $x<0.1$. The $x=0.1$ composition is unique since its $C/T$ values are proportional to $\log_{10}T$ between, at least, 1 and 10 K. We have observed deviations from this purely logarithmic behavior for values of $x$ smaller and larger than 0.1. Data shown in Fig. 1 correspond to these temperatures only when the lattice specific heat does not exceed the specific heat due to electronic degrees of freedom. Therefore, anticipated small systematical errors in the lattice specific-heat determination do not have any significant effect on the temperature dependence of $C/T$.

The trends found in $C/T$ are even more apparent in the magnetic-susceptibility results. Figure 2 shows the magnetic susceptibility versus $\log_{10}T$ for the same concentrations which are presented in Fig. 1. These measurements were performed at a field of 1 kG, with the exception of $x=0.15$, which was investigated at 100 G, and at temperatures between 1.8 and 20 K (approximately a decade of values). The susceptibility of $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ can be satisfactorily approximated by a logarithmic function of temperature. On the other hand, clear deviations from such a temperature variation are found for other in-

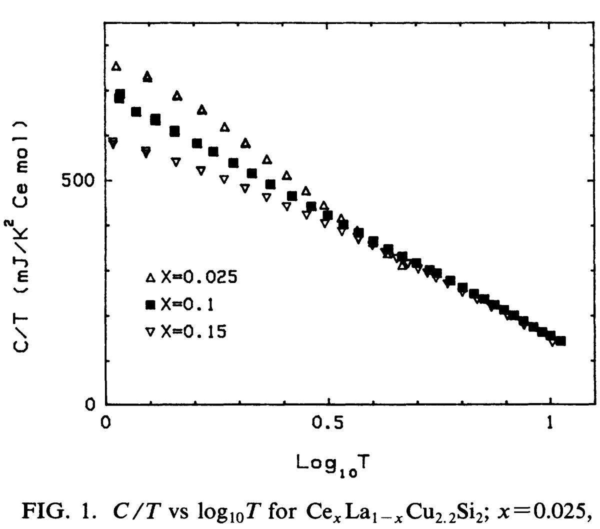

FIG. 1. $C/T$ vs $\log_{10}T$ for $Ce_xLa_{1-x}Cu_{2.2}Si_2$; $x=0.025$, 0.1, and 0.15.

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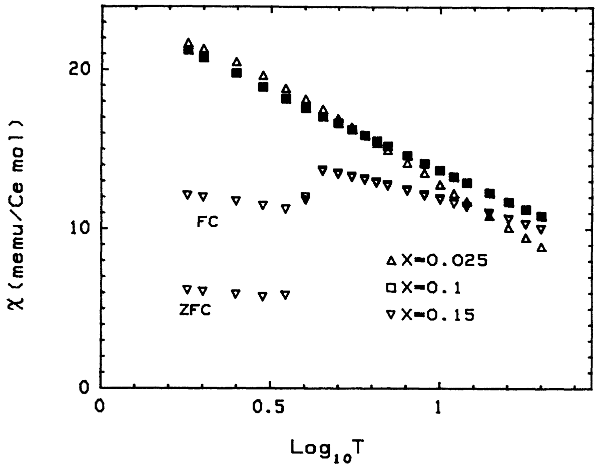

FIG. 2. $\chi$ vs $\log_{10}T$ for $Ce_xLa_{1-x}Cu_{2.2}Si_2$; $x=0.025$, 0.1, and 0.15. Measurements were performed at $H=1$ kG for $x=0.025$ and $x=0.1$, and at $H=100$ G for $x=0.15$. Points labeled ZFC (FC) correspond to the zero-field-cooled (field-cooled) $\chi$.

vestigated concentrations, the most spectacular ones for $x=0.15$, to which we will return later.

The logarithmic temperature variations of both $\chi$ and $C/T$ have been predicted for the two-channel magnetic Kondo effect.[11,12] Electrical resistivity for this model has also been calculated;[13] $\rho$ should be proportional to $T^{1/2}$. Such a temperature dependence of $\rho$ is somewhat compatible with our data (Fig. 3), but only over a limited range of temperatures between 3 and 11 K. We believe that this dependence is accidental and is due to a crossover between a logarithmic temperature variation at temperatures higher than 8 K (inset of Fig. 3) and a linear variation at temperatures lower than 5 K (Fig. 3).

The two-channel magnetic Kondo effect interpretation is clearly inconsistent with the observed magnetic-field dependence of the specific heat (Fig. 4). Small and moderate magnetic fields are expected to enhance the specific heat at temperatures small with respect to $T_K$. An increase of $C$ with $H$ is a consequence of the removal of the residual entropy which exists at $H=0$. Although shallow maxima in $C$ versus $H$ isotherms are, indeed, present in our data, both the values of these increases and the fields at which maxima occur are, by an order of magnitude, smaller than the corresponding values found in a numerical solution[12] of the problem. On the other hand, when the fields corresponding to the maxima ($H_{\max}$) are plotted versus temperature, then a straight line drawn through these points passes the $H_{\max}$ axis at $T$ close to 0 K. This finding, presently based on three points only, suggests a magnetic anomaly in the vicinity of the $T=0$ K (at zero field).

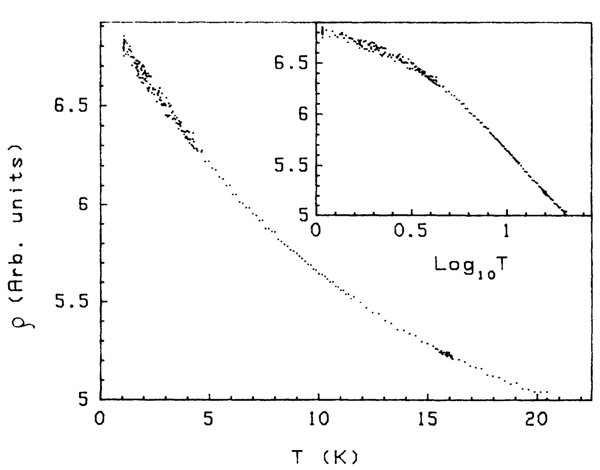

FIG. 3. Resistance ($\rho$) vs temperature for $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$. The inset shows $\rho$ vs $\log_{10}T$.

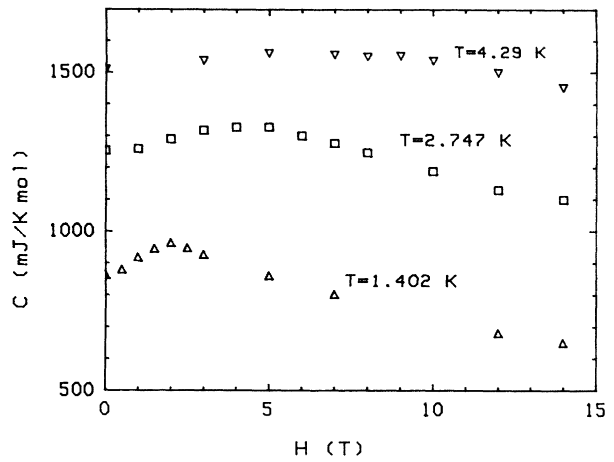

FIG. 4. Magnetic-field dependence of $C$ for $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ at $T=1.402$, 2.747, and 4.29 K.

The logarithmic temperature dependence of $C/T$ for U-based alloys have been variously associated with a two-channel Kondo effect[1] or with fluctuations of some order parameter having a critical point at $T=0$ K.[2] All experimental results that we are aware of, are consistent with a magnetic nature of the phenomenon. A small modification of the chemical composition for each of the following alloys (having $C/T\propto\log_{10}T$), $U_{0.2}Y_{0.8}Pd_3$,[1] $UCu_{3.56}Pd_{1.5}$,[3] $U_{0.1}Th_{0.9}Ni_2Al_3$, $U_{0.1}Pr_{0.9}Ni_2Al_3$,[4] can lead to a magnetic state showing some spin-glass features. Similarly, such a magnetic state can be achieved in the present system via alteration of its stoichiometry. The susceptibility of $Ce_{0.15}La_{0.85}Cu_{2.2}Si_2$ exhibits an unusual magnetic behavior at magnetic fields smaller than 1 kG (Fig. 2). There is a discontinuous drop in $\chi$ at $T$ about 4.5 K. The value of this discontinuity depends on the magnetic field and magnetothermal history of the sample. No corresponding signature has been detected in the specific heat investigated on the same sample at $H=0$ (earth's field, Fig. 1) nor at $H=100$ G. Such a behavior points to a nonequilibrium nature of the low-temperature properties in $Ce_{0.15}La_{0.85}Cu_{2.2}Si_2$.

$Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ is the first Ce-based alloy which clearly exhibits non-Fermi-liquid-like properties at low temperatures. Interestingly, similar exotic behavior has been found for other systems related to heavy-fermion superconductors: $U_xTh_{1-x}Ru_2Si_2$ ($x \leq 0.07$),[5] $U(Pt_{0.95}Pd_{0.06})_3$,[6] $U_{0.9}Pr_{0.1}Ni_2Al_3$, $U_{0.9}Th_{0.1}Ni_2Al_3$.[4]

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