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Low temperature resistivity of Ce-La-Th under pressure

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The low temperature resistivity of $Ce_{0.9-x}La_xTh_{0.1}$ alloys is known to vary as $\rho_0 + \alpha T^2$. We have investigated the variation of ρ_0 and α with pressure for several concentrations x. An unusually strong nonlinear decrease of the residual resistivity with pressure occurs; the magnitude of the decrease is an order-of-magnitude larger than in the isostructural nonmagnetic alloy $La_{0.8}Th_{0.2}$. The temperature coefficient $\alpha(P)$ also decreases strongly. These results are in qualitative accord with recent theories of the resistivity of disordered valence fluctuation compounds.

I. INTRODUCTION

In a recent article[1] we reported the observation of a novel pressure-temperature-alloy parameter (P-T-x) phase diagram for the γ-α transition in $Ce_{0.9-x}La_xTh_{0.1}$ alloys. We showed that the general features of the phase diagram follow qualitatively from a free-energy functional which incorporates in an essential way the known Fermi-liquid behavior of the 4f-spin system. In particular the Fermi-liquid temperature T_{FL} varies rapidly as the volume changes in the vicinity of the phase transition. In this paper, we examine the resistivity $\rho(T,P)$ at temperatures which are sufficiently low that the variation of ρ with temperature can be viewed as an intrinsic property of the α phase at constant T_{FL}. In earlier work on the same alloys at ambient pressure, Grier and Parks[2] demonstrated that the resistivity varies as $\rho_0 + \alpha T^2$ and reported a striking variation of the residual resistivity ρ_0 with alloy parameter x. Here we will report the pressure variation of ρ_0 and α for several values of x and will discuss our results in the context of recent theories of the transport behavior of disordered valence fluctuation compounds.

II. EXPERIMENTAL RESULTS

Experimental techniques were reported in the earlier paper[1]; here we report only results. We note, however, that the geometry of our samples did not allow determination of the absolute value of the resistivity to better than about 25%; hence, there is some minor disagreement with the absolute values reported by Grier and Parks.[2]

In Fig. 1 we exhibit the low-temperature resistivity of $Ce_{0.8}La_{0.1}Th_{0.1}$ at several pressures in such a way as to demonstrate that the resistivity varies as $\rho_0 + \alpha T^2$. We obtained comparable data for $x = 0.14$ (Fig. 2) and for $x = 0.11$ and 0.17 (not shown here). In all cases the temperature interval over which the T^2 law is obeyed increases initially with pressure. For the higher-pressure data the region of T^2 behavior is followed at higher temperature by an interval where ρ varies linearly with temperature; this in turn is followed by the phase transition. Another tendency (reported earlier by Grier and Parks[2]) is that for $x > 0.14$ the low-pressure data varies less strongly than quadratically. This can be seen for $x = 0.14$ in Fig. 2 and was also found to be true for $x = 0.17$. For higher pressures in the same samples, T^2 behavior is observed. The deviation from T^2 behavior observed in Fig. 2 at the lowest temperatures is, we believe, an extrinsic effect, perhaps due to the presence of regions of untransformed γ-cerium. Inclusion of this low temperature data leads to a power law $\rho_0 + \alpha T^n$ with $n > 2$.

Both the residual resistivity ρ_0 and the temperature coefficient α decrease rapidly with pressure. For all four concentrations studied, ρ_0 decreases to 60% of its $P = 0$ value on pressurizing to 10 kbar, while α decreases by nearly a factor of 10. This is shown in Figs. 3 and 4 for $x = 0.10$ and 0.17 respectively. By way of contrast, we show in Fig. 3(a) the resistivity data at similar pressures for the nonmagnetic al-

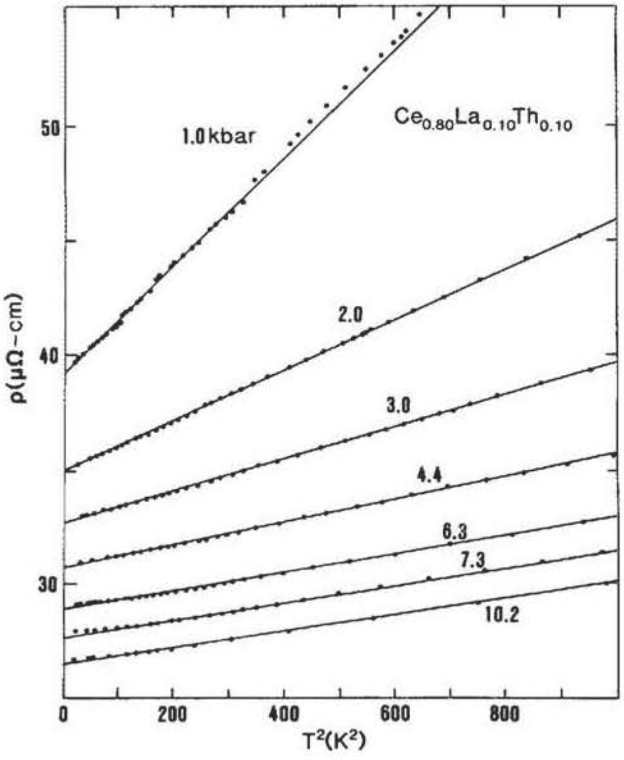

FIG. 1. The resistivity of $Ce_{0.8}La_{0.1}Th_{0.1}$ plotted vs the square of temperature for seven different pressures.

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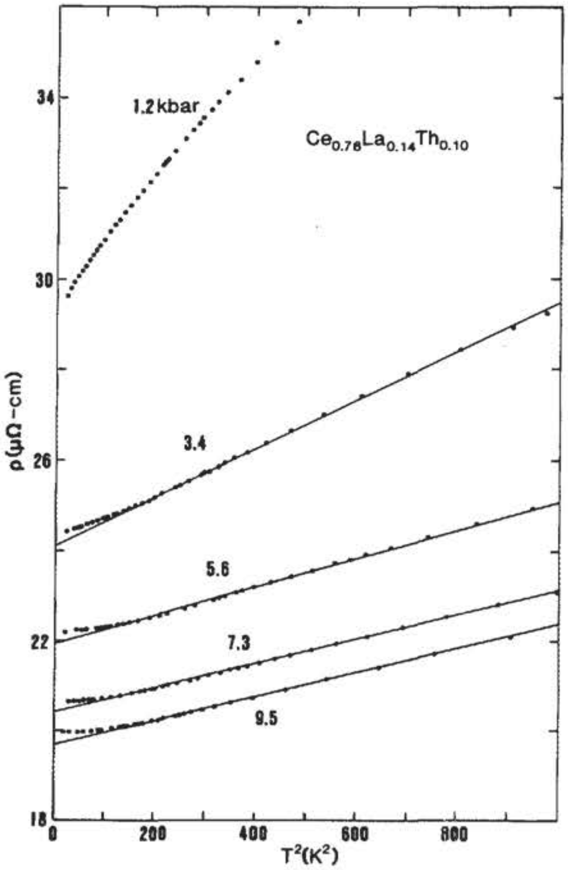

FIG. 2. The resistivity of $Ce_{0.76}La_{0.14}Th_{0.1}$ plotted vs the square of the temperature for five different pressures.

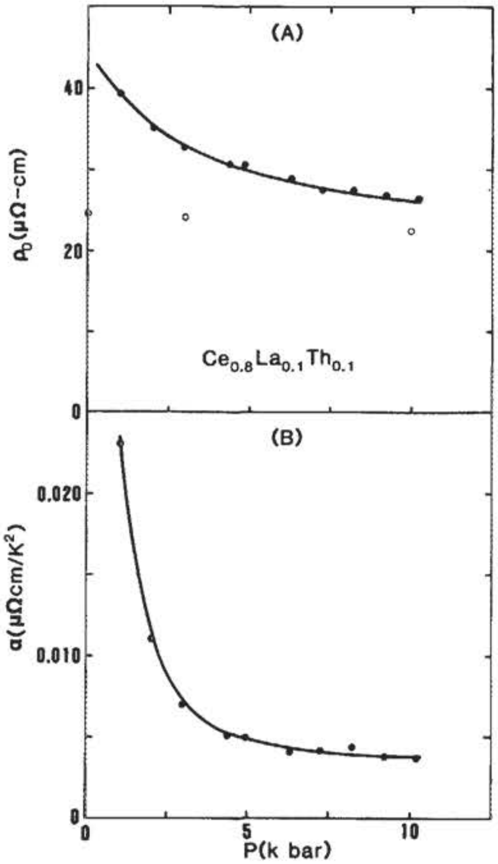

FIG. 3. (a) The residual resistivity of $Ce_{0.8}La_{0.1}Th_{0.1}$ (solid circles) and of $La_{0.8}Th_{0.2}$ (open circles) as a function of pressure and (b) the temperature coefficient α.

loy $La_{0.8}Th_{0.2}$. For this system, ρ_0 decreases by a much smaller factor (10%) between $P = 0$ and 10 kbar; and in contrast to the cerium alloys, the decrease is linear. (For $Ce_{0.8}La_{0.1}Th_{0.1}$, between 1 and 10 kbar the residual resistivity varies as P^{-6}.)

III. DISCUSSION

The strong nonlinear decrease of the residual resistivity with pressure is quite unusual for an alloy. The large magnitude of the effect (relative to the case of isostructural but nonmagnetic lanthanum alloys) suggests that the residual scattering is not simply due to the nonmagnetic solutes but involves the cerium 4*f* electrons in an essential way.

There is general agreement that the ground state of valence fluctuation metals can be described as a Fermi liquid where the conduction electrons scatter from renormalized *f* resonances whose energy scale is then T_{FL}. In a perfect lattice the resonant potential is uniform from site to site and the resistivity vanishes at $T = 0$. Finite resistivity arises from fluctuations in the resonant potential.[3,4] These can exist already at $T = 0$ due to alloy disorder and can result in a very large resistivity for two reasons.[3,5,6] First, each solute atom can cause deviations in the resonant potential at many surrounding sites: at the very least, each near neighbor is affected, and more distant neighbors can be affected if the impurity gives rise to long range strain fields. Secondly, the scattering cross section depends on the 4*f* spectral density at the Fermi level, which is large due to the resonance.

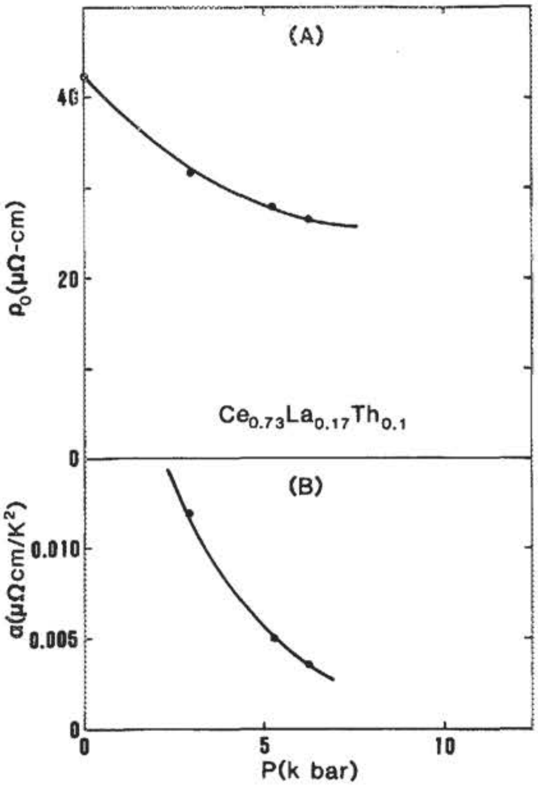

FIG. 4. A plot for $Ce_{0.73}La_{0.17}Th_{0.10}$ of (a) the residual resistivity, and (b) the temperature coefficient α as functions of pressure.

Ramakrishnan[6] hypothesizes that if the resulting disorder is sufficiently great, the conduction electrons will then scatter from a set of decoupled resonances. Utilizing the Friedel sum rule he argues that the residual resistivity will saturate to a maximum value which varies quadratically with the 4f occupation number n_f. Mihalisin and coworkers[7] have attempted to extract valences $(4 - n_f)$ in this way from residual resistivities, demonstrating strong correlations between diverse experimental quantities (lattice constants, susceptibilities, specific heat, and resistivities) consistent with such an analysis; however, the valences so obtained do not appear to agree with those obtained from x-ray absorption measurements.[8]

For temperatures greater than $T_{\rm FL}$ the conduction occurs in states away from the resonance. For strongly disordered systems the resistivity should then decrease with increasing temperature.[3] In the opposite limit of perfect order the resistivity vanishes at $T = 0$ and increases initially with temperature. The increase is due to fluctuations in the resonant potential which either can be directly excited[4,9] or, if the local resonance is coupled to lattice strain fields, can be created by thermal excitation of phonons.[6,10] For $T > T_{\rm FL}$ the resistivity begins to decrease for the reason cited above. This combination of events can lead to a resistivity maximum in pure systems, as observed in many Ce and Yb compounds. It also suggests that the initial increase of $\rho(T)$ should scale with some inverse power of $T_{\rm FL}$.

The results exhibited here for CeLaTh alloys can be understood qualitatively in these terms. The large residual resistivity arises from disordering of the 4f resonance created by the La and Th impurities. The rapid decrease of ρ_0 with pressure indicates a decreasing 4f spectral density at the Fermi level, consistent with the expected decrease in n_f with pressure. The disorder is not total, however, as indicated by the positive temperature coefficients. The large decrease in α reflects the expected broadening of the resonance (i.e., increase in $T_{\rm FL}$) with pressure,[1] with α varying as some inverse power of $T_{\rm FL}$.

Based on lattice constants, neutron linewidths[11] and x-ray absorption[12], we expect $n_f \approx 1$ at $P = 0$ for all four concentrations, and $n_f \approx 0.8$ at 10 kbar. For $x = 0.10$ at $P = 0$ it is known[11] that $T_{\rm FL} \approx 200$ K; at 10 kbar we expect $T_{\rm FL} \approx 800$ K.[8] These numbers are consistent with the observed decrease in ρ_0 [$\rho_0(10\ \text{kbar}) \approx 0.6\rho_0(P = 0)$] if $\rho_0 \propto n_f^2$. Given the sixfold decrease in α, they are consistent with a variation $\alpha \propto 1/T_{\rm FL}^n$ with $1 < n < 2$. (It would be interesting to directly observe the variations in n_f and $T_{\rm FL}$ by studies of the x-ray absorption and neutron linewidths at high pressure.) As for the observed increase in ρ_0 with x at $P = 0$ observed by Grier and Parks,[2] in our view this reflects increasing disorder, as opposed to changes in the valence.

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