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Kondo Effect in $La_{1-x}Ce_x$ Alloys under Pressure*

K. S. Kim and M. B. Maple

Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92037

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The low-temperature electrical resistance of $La_{1-x}Ce_x$ alloys in both the fcc and the dhcp phase has been measured under pressure. Between normal pressure and 19 kbar the resistance always exhibited a minimum at T_{min}, which initially increased slightly under pressure, but remained constant above 7 kbar. The resistance $R(T)$ for $T < T_{min}$ varied as $\ln T$ down to the superconducting transition temperature T_c or the limiting measuring temperature (1.3°K). The slope, $-dR(T)/d \ln T$, varied appreciably and nonmonotonically with pressure; the relationship between the depression of T_c and $-dR(T)/d \ln T$ as a function of pressure is discussed.

Previously we reported minima in the variation of the superconducting transition temperature (or maxima in the pairbreaking parameter) of $La_{3-x}Ce_xIn$ [1] and $La_{1-x}Ce_x$ [2] alloys with pressure. For the $La_{1-x}Ce_x$ alloys at pressures above 100 kbar, the depression $\Delta T_c = T_{c0} - T_c$ is more than an order of magnitude smaller than at maximum pair breaking (~15 kbar) and at least five times smaller than at normal pressure. Here T_{c0} is the superconducting transition temperature of the host metal and T_c is that of the alloy. From this it was inferred that the Ce $4f$ level moves toward the Fermi level upon the application of pressure, giving rise to an initial increase of $|J_{eff}|$, the conduction electron-impurity spin exchange coupling strength, and at sufficiently high pressure, to a transition of the Ce impurities from a magnetic to a nonmagnetic state.

Demagnetization of the Ce impurities was suggested[2] to proceed within the context of the Friedel-Anderson model.[3] The spin-up and spin-down sublevels, split below and above the Fermi level by intra-atomic Coulomb repulsion at low pressure, become degenerate and nonmagnetic at high pressure when the spin-up sublevel begins to significantly overlap the Fermi level. On the other hand, it has been suggested that a continuous increase of the Kondo temperature (T_K) with pressure could provide an alternative explanation for the pair-breaking maxima.[1,4] Both Zuckermann[5] and Müller-Hartmann and Zittartz[4] (MZ) have shown that the depression of T_c as a function of $\ln T_K/T_{c0}$ exhibits a maximum which occurs, in the MZ calculation, when $T_K \sim 12 T_{c0}$. In an attempt to determine how the Kondo temperature of $La_{1-x}Ce_x$ alloys depends upon pressure, and hence decide whether a magnetic-nonmagnetic transition or a continuous increase of T_K is responsible for the maximum and subsequent decrease in pair breaking with pressure, we have measured the low-temperature electrical resistance of $La_{1-x}Ce_x$ alloys under pressure to ~19 kbar.

Samples of $La_{1-x}Ce_x$ were prepared by melting the constituents under argon in a conventional arc furnace. The resultant ingots were then converted to the dhcp phase by cold-rolling them into foils ~0.1 mm in thickness, which were subsequently annealed in vacuum at 200°C for 3 h. To obtain the fcc phase, unannealed cold-rolled foils were heat treated in vacuum at 600°C for 10 h and then rapidly quenched in water. The agreement of the superconducting transition temperatures with the previous results[2] indicated that the right phases were obtained. A Be-Cu clamp was used to generate pressures up to 19 kbar and a Teflon bucket with a Be-Cu cap was used to contain the pressure transmitting liquid (1:1 mixture of isoamyl alcohol and n-pentane), the sample, the leads thereof, and a superconducting Pb manometer. A detailed description of the pressure seal is given elsewhere.[6] Leads were attached to the samples by spot welding and the resistance was measured by means of a standard four-lead dc technique.

In Figs. 1 and 2 are shown the curves of resistance versus temperature $[R(T)]$ measured at different pressures for fcc (2-at.% Ce) and dhcp (3-at.% Ce) $La_{1-x}Ce_x$ alloys, respectively. Although the accessible temperature range extended from 12°K down to 1.3°K, the normal state resistance of the 2-at.% Ce alloy could be measured to the lowest temperature only at 11 kbar since the sample became superconducting above 1.3°K at all other pressures. For the 3-at.% Ce alloy the resistance was measured down to 1.3°K for all applied pressures (except for normal pressure) without interference of the superconducting transition. For $T<T_{\min}$, the temperature dependence of $R(T)$ is, in all cases, nearly linear in $\ln T$ above 1.3°K or the superconducting transition temperature (marked by the sharp resistance drop). The slope $|dR(T)/d\ln T|$ distinctly increases with pressure, becoming more than twice as large at ~11 kbar than at normal pressure for both samples; it reaches a maximum near 14 kbar, beyond which it decreases. The depression of the transition temperature (or the pair-breaking parameter) behaves similarly under pressure. It increases initially, attaining its maximum value at ~14 kbar, above which it also decreases, in agreement with the previously reported results.[2] The changes of $|dR(T)/d\ln T|$, ΔT_c, and $T_{\min}$, normalized to their respective values at normal pressure, are shown in Fig. 3. $T_{\min}$ is ~6.5°K for fcc $La_{1-x}Ce_x$ (2-at.% Ce) and ~6°K for dhcp $La_{1-x}Ce_x$ (3-at.% Ce) at normal pressure, in good agreement with the results of Sugawara and Eguchi[7]; $T_{\min}$ initially increases slightly with pressure but remains constant above ~7 kbar.

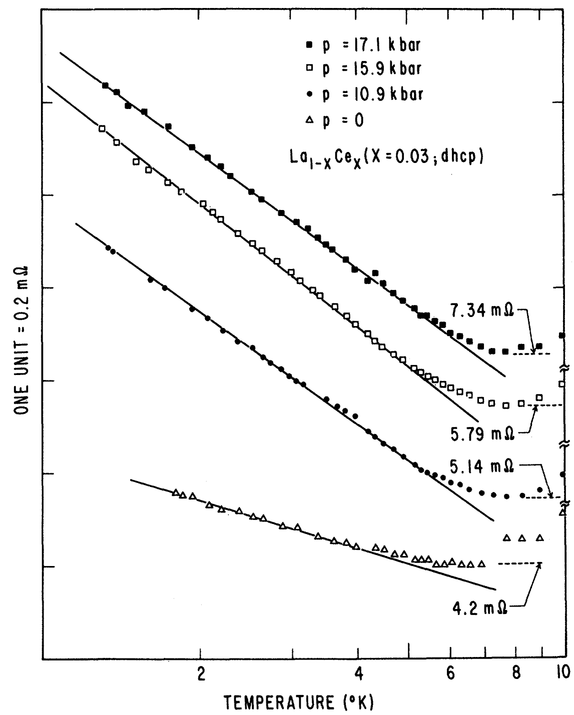

FIG. 2. Resistance of dhcp $La_{1-x}Ce_x$ (3 at.% Ce) at different pressures.

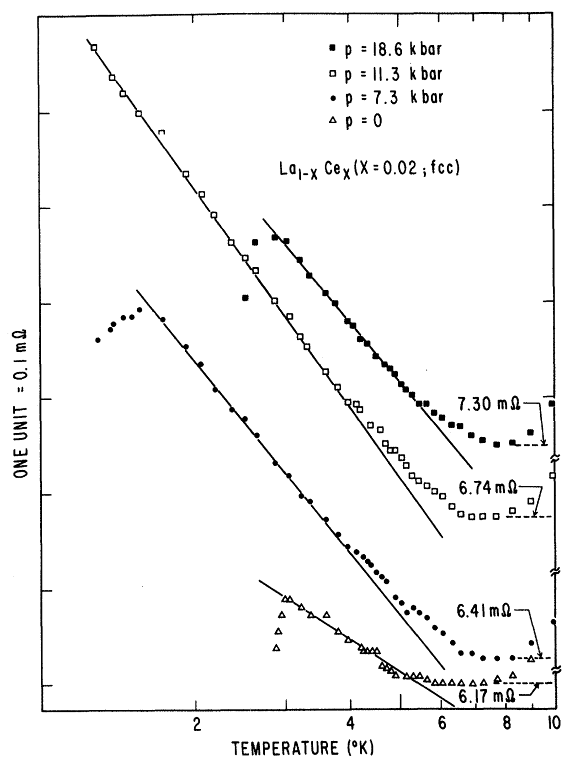

FIG. 1. Resistance of fcc $La_{1-x}Ce_x$ (2 at.% Ce) at different pressures.

The resistance of a metal containing magnetic impurities is usually approximated by $R=R(\text{host})+R(\text{impurity})$, where $R(\text{impurity})$ is the sum of R_V, the resistance due to scattering of conduction electrons by the impurity potential and R_J, the resistance due to the exchange interaction (J_{eff}) between impurity and conduction electron spins. R_J, in Suhl and Wong's result,[8] is dependent on the potential V, whereas, for example, in Abrikosov's[9] it is independent of V.

It is interesting to note that both $|dR(T)/d\ln T|$ and ΔT_c exhibit maxima at almost the same pressure. In terms of Kondo's original expression,[10] although it is valid only to third order in J_{eff} and for $T\gg T_K$, $dR_J/d\ln T$ is proportional to $N^2(0)J_{\text{eff}}^3$, where $N(0)$ is the density of states at the Fermi level. If $R(\text{host})$ is nearly independent of temperature for $T<T_{\min}$, then an increase of $|dR(T)/d\ln T|$ would imply a corresponding increase of $|J_{\text{eff}}|$, as inferred previously from the increase of ΔT_c ($\sim N(0)J_{\text{eff}}^2$ in the Born approximation[11]) with pressure. Moreover, if the decrease in pair-breaking signals the onset of a magnetic-non-magnetic transition, such a transition would intuitively lead to a decrease of $|dR(T)/d\ln T|$ since the Kondo resistance anomaly is a phenomenon of magnetic origin. From the ratio of slopes $|dR(T)/d\ln T|$ and Andres's

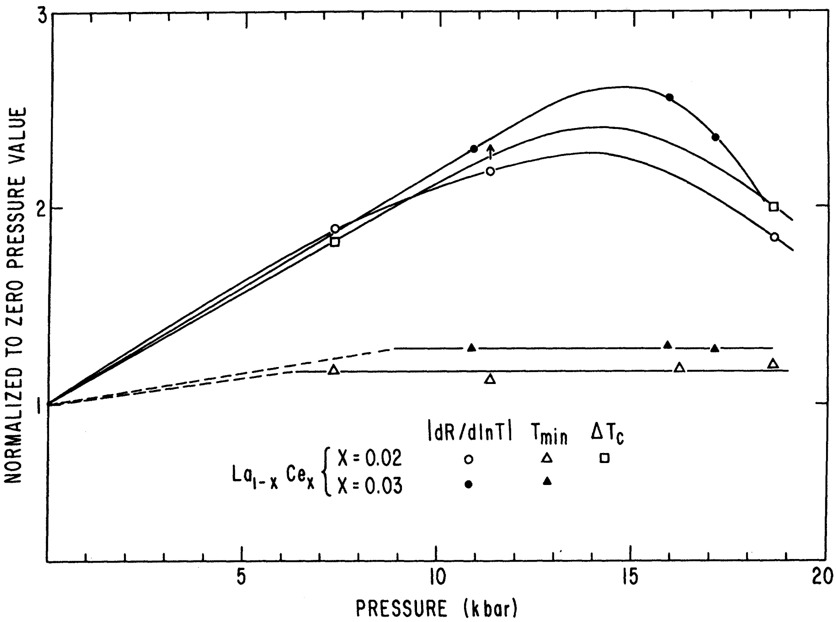

FIG. 3. $|dR(T)/d\ln T|$, ΔT_c, and T_{min} for fcc $La_{1-x}Ce_x$ (2 at.% Ce) and dhcp $La_{1-x}Ce_x$ (3 at.% Ce), normalized to their respective values at normal pressure.

value for $d\ln N(0)/d\ln V \sim -2$ from the low-temperature thermal expansion of pure La,[12] we find $|J_{eff}|$ at 11 kbar to be about 1.2 times larger than at normal pressure. Once T_K has been determined at normal pressure, a crude estimate of its initial increase with pressure may then be obtained from the relation $T_K \sim T_F \exp[-1/N(0)|J_{eff}|]$ where T_F is the Fermi temperature.

A reasonable estimate of T_K at normal pressure would be the temperature at which R_J is half of the low-temperature saturation value, i.e., its unitarity limit.[8] However, since R(impurity) should start to deviate from linearity in $\ln T$ at $T \lesssim T_K$, the observation of only $\ln T$ behavior does not allow one to estimate T_K. Sugawara and Eguchi[13] have shown from the absence of a peak in the thermoelectric power (measured above 7°K), that T_K for dilute $La_{1-x}Ce_x$ alloys is certainly lower than 7°K and probably much lower since the resistivity is still linear in $\ln T$ down to 0.4°K.

Another way of estimating T_K is from the MZ theory.[4] MZ have calculated the depression of T_c ($\Delta T_c = T_{c0} - T_c$) in terms of T_K/T_{c0} solving exactly the scattering amplitudes within the Nagaoka-Suhl approach to the Kondo problem. According to their theory, for $J_{eff} < 0$, ΔT_c first increases with $\ln T_K/T_{c0}$, reaching a maximum at $T_K/T_{c0} \sim 12$, beyond which it then decreases. Since $N(0)\Delta T_c$ is directly related to T_K/T_{c0} in the MZ theory, from their result we estimate T_K at normal pressure to be $\sim$0.6°K, using the measured ΔT_c and $N(0) = 2.44$ states/eV atom as determined from the γT term of the specific heat at low temperature.[14] From ΔT_c at 11 kbar we obtain, again using the result of MZ, the ratio $T_K(11\text{ kbar})/T_K(0) \sim 10$. This value is close to the value $T_K(11\text{ kbar})/T_K(0) \sim 8$, estimated from the change of the slope $|dR(T)/d\ln T|$, assuming $T_K \sim 0.6$°K and $T_F \sim 8\times 10^4$°K,[15] although considering the exponential relation between T_K and $|J_{eff}|$ and the approximations involved, the agreement is not to be taken too seriously. Neglecting the effect of potential scattering, the decrease of $|dR(T)/d\ln T|$ beyond 14 kbar suggests a magnetic-nonmagnetic transition; for if T_K were to continue to increase, one would expect a corresponding increase in slope with the resistivity saturating eventually at lower temperatures.

Our observations can be qualitatively interpreted in terms of the calculations of Suhl and Wong[8] in which R_J depends on V as well as J_{eff}. Inspection of their curves[16] suggests that the insensitivity of T_{min} to pressure, the continuous increase of $R(T_{min})$ and the increase of $|dR(T)/d\ln T|$ may be explained by allowing V and $|J_{eff}|$ to increase simultaneously with pressure. The higher-pressure region where $|dR(T)/d\ln T|$ decreases may correspond to a decrease or a slower increase of $|J_{eff}|$ relative to the increase of V. No theory in which R_J is independent of V can explain these features.

In conclusion, the maximum depression of T_c is readily explained by a pressure-induced magnetic-nonmagnetic transition of the Ce impurities, which leads in a natural way to the decrease of the slope $|dR(T)/d\ln T|$ observed above $\sim$14 kbar as the Ce begins to demagnetize. The alternative suggestion, that the maximum depression is due to a continuous increase of T_K, seems unlikely since this would require a special relationship between the pressure dependences of J_{eff} and V in order to explain the decrease of $|dR(T)/d\ln T|$ and yet still allow T_K to increase continuously. Moreover, from the MZ calculation, T_K would then have to attain a value $\sim 10^6$ °K to account for the observed reduction of ΔT_c at pressures $\gtrsim 100$ kbar, which seems quite unphysical. So far in our discussion we have neglected the possibility of anomalous behavior of $N(0)$ under pressure, which cannot be completely ruled out.

Discussions with Professor B. T. Matthias, Dr. D. Wohlleben, and Dr. E. Müller-Hartmann are gratefully acknowledged.

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[15] This value is obtained from the free-electron model. A recent band-structure calculation for fcc La by H. W. Myron and S. H. Liu [Phys. Rev. B **1**, 2414 (1970)] gives a value very close to this.

[16] R. M. More [Ph.D. thesis, University of California, San Diego, 1968 (unpublished)] has simplified the result of Suhl and Wong (Ref. 8) by replacing the finite range interactions with δ-function interactions. The result is

$$R_V \sim \frac{2\pi V}{1+\pi^2V^2},$$

$$R_J \sim 1 - \frac{1-\pi^2V^2}{1+\pi^2V^2}\,\frac{\ln(T/T_K)}{[\ln^2(T/T_K)+4\pi^2S(S+1)]^{1/2}}.$$

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Erratum

Evaluation of the Partition Functions for Some Two-Dimensional Ferroelectric Models, M. L. Glasser, [Phys. Rev. **284**, 359 (1969)].

(1) On the right-hand side of Eq. (13), λ should be replaced by λ^{-1}.

(2) In Eq. (14) the argument of the logarithm should be

$$\Gamma(\alpha\beta/2\pi+\tfrac{3}{4})\,\Gamma(\alpha\gamma/2\pi+\tfrac{1}{4})/[\Gamma(\alpha\gamma/2\pi+\tfrac{3}{4})\,\Gamma(\alpha\beta/2\pi+\tfrac{1}{4})].$$

(3) To Eq. (23) add $=\ln|(2\mu/\pi)\cot(\pi^2/2\mu)\csc\mu|$.

(4) The right-hand side of Eq. (24) should read

$$(\tfrac{1}{8}\mu)I(\pi/2\mu, 3\mu, 5\mu).$$

(5) Equation (27) should read

$$z(0)=\ln|(2\mu/\pi)\cot\mu\cot(\pi^2/2\mu)|.$$

I wish to thank Dr. D. B. Abraham for pointing out the above simple form for Eq. (27).