PRESSURE DEPENDENCE OF THE RESISTIVITY OF CE_3Al

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Above 200C the compound Ce_3Al has a cubic (Cu_3Au) structure; below 200C it transforms to a hexagonal (Ni_3Sn) phase.[1] The susceptibility[2] in these phases is of Curie-Weiss form $\chi(T) = C/(T+\theta)$ with $\mu_{eff} = 2.52\ \mu_B$ and $\theta = 24K$. The situation is reminiscent of the γ-β transition in elemental cerium[3]; the high temperature phase being γ-like (i.e., fcc but with the Al atoms at the cell corners) and the lower temperature phase being β-like (although ordered hcp rather than dhcp). For this reason we have undertaken studies of the pressure and temperature dependence of the resistivity $\rho(P;T)$ to search for evidence of a transition to an α-Ce-like state. We find that given sufficiently high purity samples there is a structural transition near 100K which may indeed be isomorphic (i.e., involve no change in crystal symmetry as in the γ-α transition of Ce). The behavior in the low temperature phase varies rapidly with pressure: it is a heavy fermion antiferromagnet at P=0 but transforms smoothly to a nonmagnetic large Kondo temperature compound at higher pressures analogous to α-cerium.

In an earlier report[4] we exhibited $\rho(PT)$ curves for a sample made from starting materials obtained from commercial sources. We have recently repeated the measurements on a sample made from very high purity cerium obtained from the Materials Preparation Center of the Ames Laboratory. In addition, whenever possible we handled the starting materials or sample in a helium filled glove bag. (Other experimental details are given in the earlier report.[4]) The results are shown in Fig. 1. They differ from those reported earlier in one major respect: in the earlier results a broad maximum was observed in the resistivity near 100K; here an actual first order phase transition occurs. The results differ in minor respects as well; for example, the region near 10K of negative slope disappears at a higher pressure in the new sample than in the old. Furthermore, while metallographic examination and x-ray diffraction usually showed in the old samples[2,4] a trace of the cubic Cu_3Au phase at room temperature, the new samples showed no sign of the cubic phase. We thus believe that the new samples represent the truth about Ce_3Al. The

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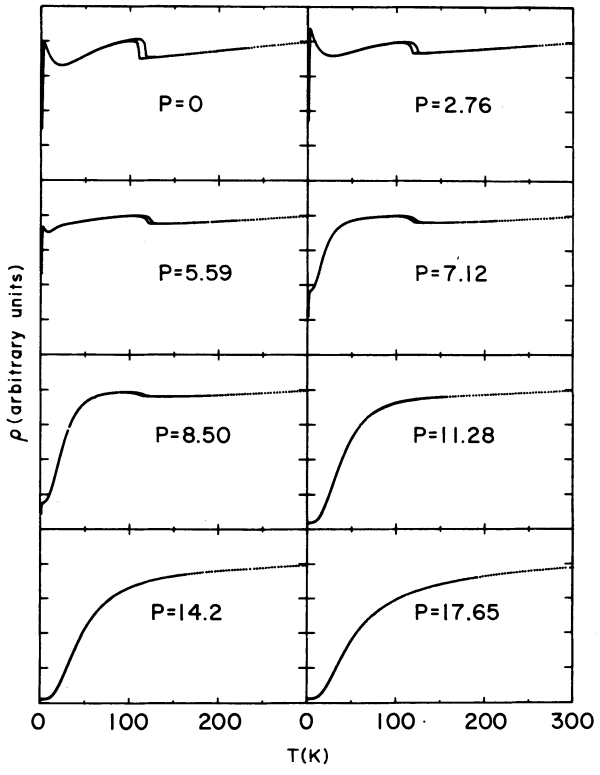

Fig. 1. Resistivity versus temperature of Ce_3Al at eight pressures (given in kbar).

most likely cause of the difference is the absence of oxygen and other interstitials in the new samples; these apparently[5] act to stabilize the cubic phase and also to suppress the phase transition.

The results shown in Fig. 1 indicate the existence of a structural transition near 100K in Ce_3Al. The resistivity in the vicinity of the phase transition is enlarged in Fig. 2. The height of the discontinuity decreases as the pressure is raised and the magnitude of the hysteresis decreases. Above 20 kbar the discontinuity and the hysteresis have vanished. These features would follow if a critical endpoint for the first order phase transition existed in the PT plane in the vicinity of 10 kbar and 100K. Such a critical point would only be possible if the phase transition did not change the crystal symmetry -- i.e., the transition would have to be an isomorphic[3] transition between two hexagonal (Ni_3Sn) phases where only the lattice constants change at the first order transition. Such a transition would be analogous to the γ-α transition in

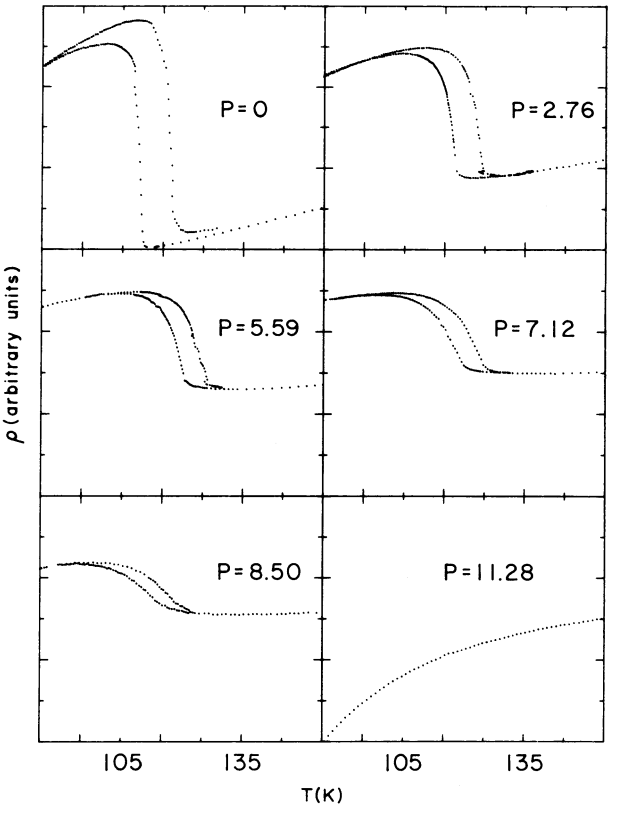

Fig. 2. Resistance versus temperature in the vicinity of the structural phase transition at six pressures (given in kbar).

cerium, but would occur within a hexagonal phase.

While the phase transition may be analogous to the γ-α transition, there are strong differences between the ground state of Ce_3Al and that of α-cerium. First of all, at low temperatures at ambient pressure the resistivity is Kondo-like ($\partial\rho/\partial T < 0$); estimating the resistance minimum as the Kondo temperature gives $T_K \approx$ 10-20K. We find that the susceptibility peaks at T_N = 2.7K, implying the existence of antiferromagnetic order. The resistivity also peaks at this temperature, dropping rapidly at lower temperatures. The Kondo temperature and large value of χ observed at T_N (0.03 emu/mole-Ce) indicate that the ordering occurs in a heavy fermion compound,[6] in a regime where $\partial\rho/\partial T < 0$.

As the pressure is raised the Kondo resistivity (negative $\partial\rho/\partial T$) decreases and disappears above P = 6 kbar. At higher pressures (P > 10 kbar) the resistivity takes on the form associated with a large T_K compound: i.e., the Kondo temperature varies rapidly with pressure in the low temperature phase, in contrast to the room temperature phase where the variation of ρ with P is negligible. At the same time the temperature and magnitude of the resistivity maximum decrease with pressure; the magnetic transition disappears for P > 10 kbar. Hence, at the highest pressures shown the resistivity shows behavior analogous to α-cerium, i.e., a nonmagnetic large T_K compound.

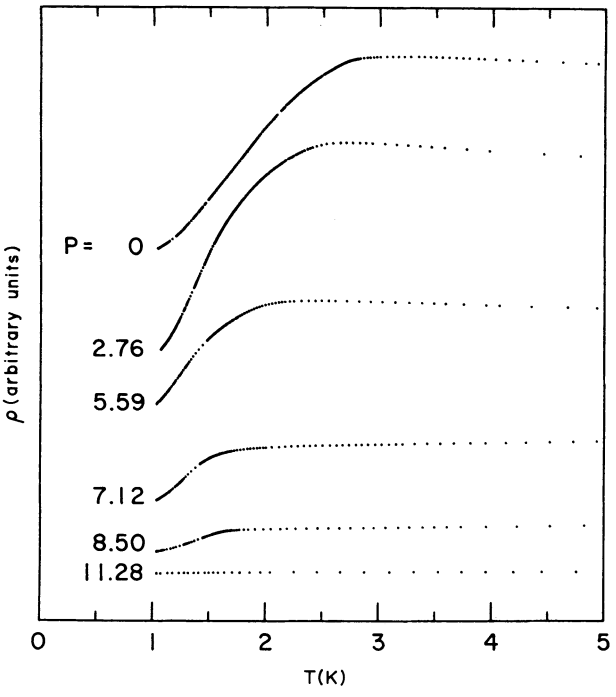

Fig. 3. Resistance versus temperature in the vicinity of the antiferromagnetic phase transition at six pressures (given in kbar). The top two curves have been shifted up for clarity.

The behavior of Ce_3Al is thus seen to be quite rich. A structural transition occurs which may be isomorphic; in the ground state the antiferromagnetic heavy fermion behavior observed at ambient pressure evolves smoothly towards nonmagnetic large T_K (slightly overweight fermion) behavior as the pressure is raised. A T = 0 magnetic/nonmagnetic transition is observed. These results -- the nature of the structural and magnetic transitions -- need to be confirmed by neutron scattering.

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