## MAGNETIC-NONMAGNETIC TRANSITIONS IN ALLOYS WITH CERIUM IMPURITIES



FIGURE 4  $|dR_m/d \ln T|$ ,  $\Delta T_c$  and  $T_{min}$  for a LaCe (2 at. % Ce) alloy (normalized to their respective values at normal pressure) to 18 kbar (reference 11).

increases slowly with pressure. Shown in Figure 5 is the resistivity vs temperature curve of an  $Y_{0.99}Ce_{0.01}$  alloy at different pressures.<sup>12</sup> The resistivity minimum, present at low pressure, disappears completely at high pressure.

## II THE THEORETICAL MODEL

It is well recognized that in the dilute alloy limit no sharp transition is expected, either as a function of temperature or pressure. The meaning of magnetic or nonmagnetic impurity states is then relative and qualitative. Keeping this in mind, we can grosso modo characterize three typical regions (on a  $T_{,p}$  plane for example):

- (i) a high temperature  $T > T_k$  ( $T_k =$ Kondo temperature) magnetic region;
- (ii) a low temperature  $T < T_k$  nonmagnetic (or condensed magnetic) region; and
- (iii) a normal nonmagnetic region.

Hence we see that a magnetic-nonmagnetic transition can occur in two typical ways: (i)  $\rightarrow$  (ii) and (i)  $\rightarrow$  (iii).

In the first case, it will be convenient to use throughout a Kondo Hamiltonian with some assumed or adjusted dependence of  $T_k$  on the pressure.

In this paper, we explore the second case. The resistivity experiments on LaCe up to 18 kbar indicate that  $T_k$  remains very small in this pressure range and that the exchange integral exhibits a maximum near 15 kbar instead of a monotonically



FIGURE 5 Resistivity versus temperature of YCe (1 at. % Ce) at various pressure up to 79 kbar (reference 12).

increasing variation as a function of pressure. In the absence of susceptibility and specific heat measurements, we have to rely on these indications from the resistivity to discriminate between cases 1 and 2 because the variation of  $T_c$  will be qualitatively similar in both instances.

We assume that the effect of pressure is to shift linearly the energy  $E_{4f}$  of the 4f level upwards with respect to the Fermi level  $E_F$ . In the absence of a good solution of the Anderson Hamiltonian throughout the region where the impurity level crosses the Fermi level, we attack this region from the left (low pressures,  $E_{4f} < E_F$ ) and from the right (high pressures,  $E_{4f} > E_F$ ). The aim is to see how far we can go from each side and to see how the parameters needed to fit the data compare with those obtained in pure Ce or in other Ce alloys. The results obtained show a rather good degree of selfconsistency; further experiments on these alloys will tell if the model encompasses the essential features.

335

In the magnetic domain below  $p_c$  ( $\simeq 30$  kbar), the 4f level is below  $E_F$  and the Anderson Hamiltonian can be reduced to an exchange Hamiltonian

$$\mathscr{H} = -\Gamma s \cdot S \tag{1}$$

describing the interaction between the conduction electron spin density s at the impurity site and the localized electron spin S. The interaction constant, as previously described,<sup>3, 5</sup> is the sum of the two terms:

$$\Gamma = \Gamma_1 + \Gamma_2 \tag{2}$$

 $\Gamma_1$  comes from the normal exchange scattering mechanism and is small, positive and nearly pressure independent. We make here the reasonable approximation that  $\Gamma_1$  is constant in the rare-earth series and take the value deduced for gadolinium impurities for which  $\Gamma_2$  is nearly zero.  $\Gamma_2$  arises from the resonant scattering mechanism, is negative, and can be obtained from the Schrieffer–Wolff transformation<sup>13</sup>

$$\Gamma_2^0 \simeq -\frac{2V_{kf}^2}{|\varepsilon|} \tag{3}$$

 $V_{kf}$  is the matrix element of mixing and  $\varepsilon(\varepsilon < 0)$  is the energy separating the 4f level and  $E_{\rm F}$ . Formula (3) is no longer valid when  $|\varepsilon|$  becomes very small, i.e.  $|\varepsilon|$  smaller than  $\Delta$ . As  $\varepsilon$  approaches zero, the the phase-shift  $\delta_v$  of the occupied 4f level varies rapidly with  $\varepsilon$ . Thus in the region of very small  $\varepsilon$ , we have to take into account the direct scattering Hamiltonian which gives the  $\varepsilon$  dependent phaseshift  $\delta_v$ .<sup>14, 15</sup> One effect of the direct scattering term is to renormalize  $\Gamma_2^0$  into an effective  $\Gamma_2$  approximately given by

$$\Gamma_{2} = \Gamma_{2}^{0} \cos^{2} \delta_{v} = \frac{2V_{kf}^{2}}{\varepsilon} \frac{\varepsilon^{2}}{\varepsilon^{2} + \Delta^{2}}$$
$$= -\frac{2V_{kf}^{2} |\varepsilon|}{\varepsilon^{2} + \Delta^{2}} \qquad (4)$$

 $\Gamma_2$  has a maximum when  $\varepsilon = -\Delta$  and is zero when  $\varepsilon = 0$ . We apply this ionic model until  $\varepsilon = 0$  although, obviously, the nearer we approach the transition, the less valid the ionic model and, in turn, the less valid formulas (2) and (4).

In the nonmagnetic domain above  $p_c$ , the 4f level is above  $E_F$  at a distance E and we use the nonmagnetic resonant states theory within the Hartree– Fock approximation. E is much larger than the half-width  $\Delta$  in the nonmagnetic domain because the Coulomb repulsion is very much larger than Eand  $\Delta$  in rare-earth metals. The total 4f density of states for the two spin directions is:

$$n_f(E_{\rm F}) = \frac{\xi}{\pi\Delta} \left(\frac{\Delta}{E}\right)^2 \tag{5}$$

and the total number of 4f electrons N is given by

$$N = \frac{\xi}{\pi} \frac{\Delta}{E} = \sqrt{\frac{\xi \Delta n_f(E_F)}{\pi}}$$
(6)

 $\xi$  is the degeneracy of the nonmagnetic 4f state which is equal to 14 if we do not take into account spin-orbit coupling. On the other hand, if the spin-orbit coupling is large relative to  $\Delta$ , as is usually the case in rare-earths, the 4f level is split in a j = 5/2 and a j = 7/2 state. For cerium, the ground state is j = 5/2 and we can consider it the only occupied state, so that  $\xi$  is equal to 6.

## **III RESULTS**

 $(1^{\circ})$  Depression of the superconducting transition temperature.

We calculate  $-dT_c/dc$  in the magnetic domain  $(T > T_k)$ . At low concentration  $-dT_c/dc$  is given by the Abrikosov–Gor'kov formula:<sup>16</sup>

$$-\left(\frac{\mathrm{d}T_c}{\mathrm{d}c}\right)_{c=0} = \frac{\pi^2}{4} n_s(E_\mathrm{F}) S(S+1)\Gamma^2 \tag{7}$$

with  $\Gamma$  given by (2) and (4):

$$\Gamma = \Gamma_1 + 2\Gamma_0 \frac{\Delta \varepsilon}{\varepsilon^2 + \Delta^2} \tag{8}$$

where

$$\Gamma_0 = \frac{2}{\pi n_s(E_{\rm F})} \tag{9}$$

We take the value  $n_s(E_F) = 4.4$  states/eV atom for the density of states of the sd band of pure lanthanum (for two spin directions) deduced from specific heat measurements. Thus,  $\Gamma_1 = 0.028$  eV is deduced from experiments on LaGd<sup>5</sup> where  $\Gamma_2 \sim 0$ ,  $\Gamma_0 =$ +0.145 eV and we assume, as usual,<sup>1</sup>  $\Delta = 0.02$  eV.

We further assume a linear variation of  $\varepsilon$  with pressure of 0.02 eV per 15 kbar and choose  $p_c$  equal to 32 kbar. Thus  $\varepsilon = -(32/15)\Delta = -0.0427$  eV at normal pressure. Using these values and equations (7), (8) and (9) we plot the theoretical curve (labeled (I)) in Figure 1. Hence, by assuming a reasonable