Intern. J. Magnetism, 1971, Vol. 1, pp. 333–340

MAGNETIC–NONMAGNETIC TRANSITIONS IN ALLOYS WITH CERIUM IMPURITIES

B. COQBLIN

Laboratoire de Physique des Solides† *Faculté des Sciences,* 91 *Orsay, France*

M. B. MAPLE‡ and G. TOULOUSE¶

University of California, San Diego, La Jolla, California 92 037, *U.S.A.*

(*Received* 17 *February* 1971)

The continuous transition of dilute metallic alloys from magnetic to nonmagnetic behaviour has been recently observed experimentally in alloys containing Cerium impurities. A brief review of these experiments, which include measurements of the resistivity and of the depression of the superconducting temperature under applied pressure, is presented in connection with the predictions of a phenomenological model. The parameters needed to fit the existing data are consistent with those obtained in pure Ce or in other Ce alloys.

I EXPERIMENTAL INTRODUCTION

It is well-known that cerium, either as a pure metal or an impurity dissolved in a non magnetic matrix, exhibits peculiar properties related to the proximity of the Ce 4f level to the Fermi level E_F.[1,2] Alloys with cerium impurities have been extensively studied in previous years[2,3] and their properties depend strongly on the relative position of the Ce 4f level and E_F; they are non magnetic when the 4f level is above E_F, and magnetic when it is below. From experiments by Smith[4] on the pressure dependence of the superconducting transition temperature T_c of **La**Ce alloys, it has been estimated[5] that the Ce 4f level lies a few hundredths of an eV below E_F at normal pressure and moves towards E_F by a hundredth of eV under a pressure of 10 kbar. Hence, **La**Ce as well as **Y**Ce have been suggested[5] as promising candidates for an experimental study of the transition from magnetism to nonmagnetism at feasibly higher pressures.

A minimum in the variation of T_c with pressure was first observed in the (**La**Ce)$_3$In system and interpreted as signalling the onset of a magnetic–nonmagnetic transition.[6] In these alloys, for a given concentration c of magnetic cerium impurities, the depression of T_c, $\Delta T_c \equiv T_{c0} - T_c$ where T_{c0} is the transition temperature of the matrix, initially increases strongly with pressure and then goes through a maximum at higher pressure. Recently, Maple *et al.*[7] have measured the variation of T_c with pressure in **La**Ce alloys to 140 kbar. They observed that T_c of pure lanthanum increases monotonically to 12°K at 140 kbar, while, for example, T_c of a $La_{0.987}Ce_{0.013}$ alloy first decreases with pressure and then goes through a minimum at 15 kbar. This behaviour is so pronounced in a $La_{0.98}Ce_{0.02}$ alloy, that there is a 'normal gap' between 5 and 15 kbar on the pressure axis where the sample is not superconducting above the lowest temperature (0.35°K) accessible to the experiment. Hence, ΔT_c shows a maximum as a function of pressure. In figure 1, we have plotted $\Delta T_c/c$ for the $La_{0.987}Ce_{0.013}$ alloy versus pressure to 140 kbar. The depression of T_c at high pressure ($\gtrsim$ 100 kbar) is more than an order of magnitude smaller than at the maximum depression at 15 kbar. Moreover, the shape of the T_c versus c curve changes with pressure, as seen in Figure 2. At low pressures, it is nearly linear with a small negative curvature, while at high pressures the curvature becomes positive and is strongest at roughly 30 kbar. From Figure 1, the magnitude of the depression of T_c is typical of a magnetic impurity (like other rare-earth impurities) at low pressure (below $\sim$ 30 kbar), while it is typical

† Laboratoire associé au C.N.R.S.

‡ Research sponsored by the Air Force Office of Scientific Research, Office of Aerospace Research, United States Air Force, under Grant No. AF-AFOSR-631-67-A.

¶ Permanent address: Laboratoire de Physique des Solides—Faculté des Sciences—91—Orsay—France.

of a nonmagnetic impurity at very high pressure (above $\sim$ 100 kbar).

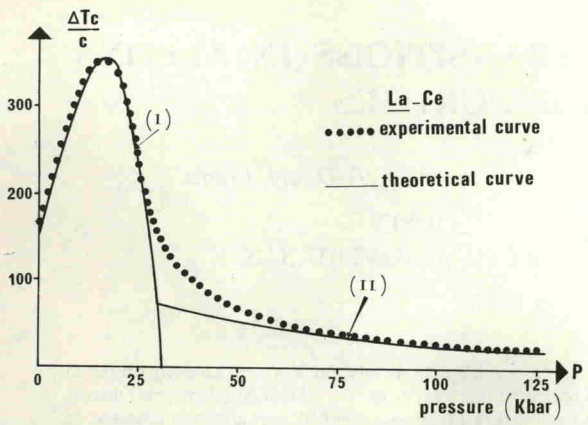

FIGURE 1 Depression of the superconducting transition temperature $\Delta T_c/c$ of a **La**Ce (1.3 at. % Ce) alloy versus pressure to 140 kbar (reference 7) compared to the theoretical curves (I and II) discussed in the text.

The preceding variation of ΔT_c (or $-\mathrm{d}T_c/\mathrm{d}c$), characteristic of the pressure induced magnetic–nonmagnetic transition in **La**Ce alloys, can also be obtained in a ternary alloy by varying the composition of the matrix.[8] This is shown in Figure 3. To the right we have plotted $(-\mathrm{d}T_c/\mathrm{d}c)_{c=0}$ vs pressure for **Th**Ce alloys[9,10] which decreases with pressure in much the same manner as for **La**Ce alloys at very high pressure; to the left we have plotted the same quantity as a function of the relative composition of the matrix, i.e., the yttrium concentration x in ternary $(\mathrm{Th}_{1-x}\mathrm{Y}_x)_{1-c}\mathrm{Ce}_c$ alloys.[8] The variation of $(-\mathrm{d}T_c/\mathrm{d}c)_{c=0}$ with x is similar to that which occurs with decreasing pressure in nonmagnetic **La**Ce alloys.

The electrical resistivity is another quantity which is expected to show a remarkable variation in the vicinity of the magnetic–nonmagnetic transition. Figure 4 shows the slope of the Kondo resistivity $|\mathrm{d}R_m/\mathrm{d}\ln T|$ (normalized to the value at zero pressure) in the temperature range where R_m is linear in $\ln T$, and the temperature of the resistivity minimum $T_{\min}$ (also normalized to the zero pressure value) vs pressure to 18 kbar for a $\mathrm{La}_{0.98}\mathrm{Ce}_{0.02}$ alloy.[11] In the same figure ΔT_c, determined from resistive transitions on the same sample when the alloy is superconducting or extrapolated when it is no longer superconducting, is shown for comparison. Experimentally, $|\mathrm{d}R_m/\mathrm{d}\ln T|$ and ΔT_c have a maximum at nearly the same pressure, while $T_{\min}$

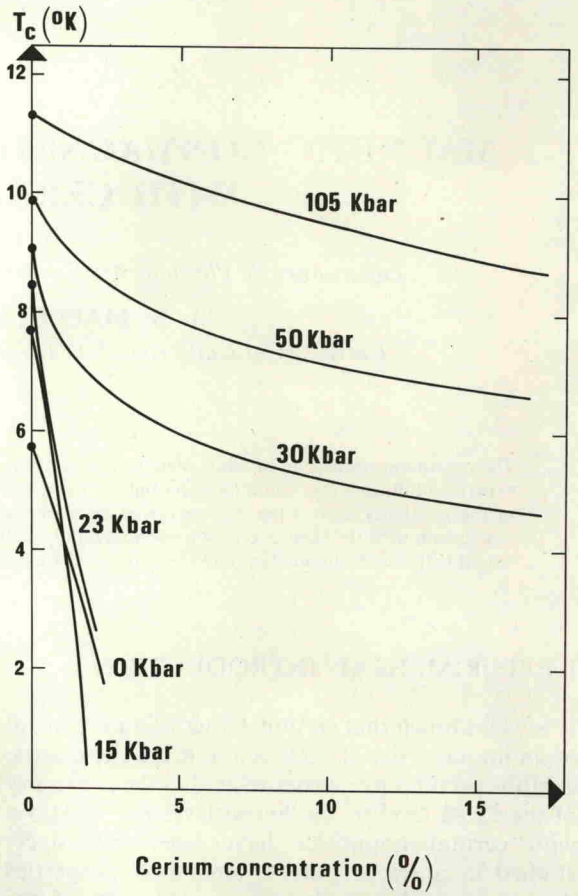

FIGURE 2 Isobaric concentration dependence of the superconducting transition temperature T_c in the **La**Ce system (reference 7).

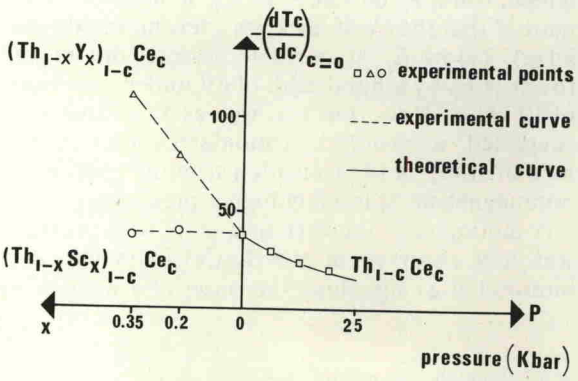

FIGURE 3 Initial depression of the superconducting transition temperature $(\mathrm{d}T_c/\mathrm{d}c)_{c=0}$ versus pressure in the $\mathrm{Th}_{1-c}\mathrm{Ce}_c$ system, and matrix composition x in the $(\mathrm{Th}_{1-x}\mathrm{Y}_x)_{1-c}\mathrm{Ce}_c$ and $(\mathrm{Th}_{1-x}\mathrm{Sc}_x)_{1-c}\mathrm{Ce}_c$ systems (reference 8).