© GORDON AND BREACH SCIENCE PUBLISHERS LTD. Printed in Norwich, England

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 $E_{\rm F}$.^{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 $E_{\rm F}$; they are non magnetic when the 4f level is above $E_{\rm F}$, and magnetic when it is below. From experiments by Smith⁴ on the pressure dependence of the superconducting transition temperature T_c of LaCe alloys, it has been estimated⁵ that the Ce 4f level lies a few hundredths of an eV below $E_{\rm F}$ at normal pressure and moves towards $E_{\rm F}$ by a hundredth of eV under a pressure of 10 kbar. Hence, LaCe as well as YCe have been suggested⁵ 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 (LaCe)₃In system and interpreted as signalling the onset of a magnetic-

¶ Permanent address: Laboratoire de Physique des Solides— Faculté des Sciences—91—Orsay—France. nonmagnetic transition.⁶ In these alloys, for a given concentration c of magnetic cerium impurities, the depression of T_c , $\Delta T_c \equiv T_{c_0} - T_c$ where T_{c_0} 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 LaCe 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 ~ 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.

B. COQBLIN, M. B. MAPLE AND G. TOULOUSE

of a nonmagnetic impurity at very high pressure (above ~ 100 kbar).



FIGURE 1 Depression of the superconducting transition temperature $\Delta T_c/c$ of a LaCe (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 -dT/dc). characteristic of the pressure induced magneticnonmagnetic transition in LaCe 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 $(-dT_c/dc)_{c=0}$ vs pressure for ThCe alloys^{9, 10} which decreases with pressure in much the same manner as for LaCe 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 $(Th_{1-x}Y_x)_{1-c}Ce_c$ alloys.⁸ The variation of $(-dT_c/dc)_{c=0}$ with x is similar to that which occurs with decreasing pressure in nonmagnetic LaCe 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 $|dR_m/d \ln T|$ (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 La_{0.98}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, $|dR_m/d \ln T|$ 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 LaCe system (reference 7).



FIGURE 3 Initial depression of the superconducting transition temperature $(dT_c/dc)_{e=0}$ versus pressure in the Th_{1-c}Ce_c system, and matrix composition x in the $(Th_{1-x}Y_x)_{1-c}Ce_c$ and $(Th_{1-x}Sc_x)_{1-c}Ce_c$ systems (reference 8).

334