EVIDENCE FOR THE PRESENCE OF THE KONDO EFFECT IN THE COMPOUND CeAl₂

K. H. J. Buschow and H. J. van Daal

Philips Research Laboratories, N. V. Philips' Gloeilampenfabrieken, Eindhoven, The Netherlands

(Received 17 June 1969)

The anomalies present in the electrical resistivity of $CeAl_2$ have been suppressed by partial replacement of Ce by Th. It is argued that this substitution influences appreciably the density-of-states function of the conduction electrons at the Fermi surface. The result of the Th substitution is thought to bring further evidence in favor of the occurrence of the Kondo effect in $CeAl_2$.

The occurrence at low temperatures of resistivity minima for the case of alloys having dilute, or in some cases also nondilute, contents of Ce as a magnetic impurity has been reported to be indicative of the formation of a spin-compensated or Kondo state.¹⁻⁶ Minima have recently also been observed for the case of dilute⁷ and nondilute⁸ Ce intermetallic compounds in which, contrary to allov systems, the Ce ions are restricted to specified lattice positions. The occurrence of spin compensation has also been suggested, on the basis of susceptibility data, for the element Ce.⁹ Current theories of the Kondo effect provide an explanation of these resistivity minima only for dilute systems.^{10,11} The Kondo temperature $T_{\rm K}$, characteristic for the onset of spin compensation, depends exponentially on the product $n(E_{\rm F})|J|$, where $n(E_{\rm F})$ denotes the density of states of the conduction electrons at the Fermi surface and Jthe (negative) effective s-f exchange coupling constant. A decrease of $n(E_F)$ and/or |J| should give rise to a marked diminution of $T_{\rm K}$. The simple exponential dependence of $T_{\rm K}$ on $n(E_{\rm F})$ is considered to be essential in explaining the data presented below, although a possible discrepancy with experiment has been reported in literature recently.¹²

In this paper the resistivity (ρ) behavior between 1.3 and 50°K is reported for the cases of CeAl, and of some ternary compounds $Ce_{1-x}Th_x$ - Al_2 and $Ce_{1-x}La_xAl_2$. The methods used in the preparation of the samples and in the measurement of the resistivity have been described in Ref. 8. From magnetic susceptibility measurements we found indications that replacement in RAl_2 (R = trivalent nonmagnetic rare-earth element) of R by tetravalent (nonmagnetic) Th leads eventually to a pronounced decrease of $n(E_{\rm F})$. Xray diffraction has shown that at a certain Th concentration the crystal structure changes from the cubic $MgCu_2$ type (RAl_2) to the hexagonal AlB₂ type (ThAl₂). The pronounced decrease of $n(E_{\rm F})$ mentioned can indeed be expected just below the value for *x* corresponding to the structure change. For $Ce_{1-x}Th_xAl_2$ the structure change

occurs approximately at x = 0.45. Ce_{1-x}La_xAl₂ remains cubic at all values of x. Resistivity data for cubic compounds are shown in Fig. 1. For the compounds $Ce_{1-x}Th_xAl_2$, ρ decreases rapidly when the temperature is lowered below approximately 5°K. Data for the magnetic susceptibility suggest that these compounds are antiferromagnetic with Néel temperatures between 3.5 and 6° K. It is concluded that the decrease of ρ below 5°K is connected with the onset of magnetic ordering. For the compounds $Ce_{1-x}La_xAl_2$ magnetic ordering seems to occur at temperatures lower than those considered in this investigation. The difference between ρ and residual resistivity (ρ_r) as a function of temperature for the compounds $Ce_{1-x}Th_xAl_2$ is presented in Fig. 2.

The data presented in Figs. 1 and 2 seem to provide additional evidence for the presence of the Kondo effect in CeAl₂ above the Néel tempera-



FIG. 1. Resistivity (ρ) as a function of temperature for $\text{Ce}_{1-x} \text{La}_x \text{Al}_2$ and $\text{Ce}_{1-x} \text{Th}_x \text{Al}_2$ compounds, all having the cubic MgCu₂ structure.



FIG. 2. The measured resistivity (ρ) minus the residual resistivity (ρ_r) for the compounds $\text{Ce}_{1-x}\text{Th}_x\text{Al}_2$ as a function of temperature. With regard to the cases of 5 and 10% Th, below 10°K the upper curve is representative of 5% Th.

ture:

(1) The resistivity minimum disappears if in CeAl₂, Ce is replaced by Th to the extent of more than 10%. This can be expected because this substitution leads eventually to an appreciable decrease of $n(E_{\rm F})$ and thus to a drastic reduction of $T_{\rm K}$, as well as to a substantial reduction of the normal spin-disorder resistivity.

(2) The maximum value of ρ at the ordering temperature ($\approx 5^{\circ}$ K) decreases far more rapidly than proportional to the dilution in CeAl₂ of Ce with Th. This can be understood because for CeAl₂, having a relatively large value for $n(E_{\rm F})$, the normal spin-disorder resistivity is relatively large and moreover the Kondo resistivity appreciable.

(3) For the $Ce_{1-x}La_xAl_2$ compounds our data together with those of Ref. 7 $(1-x \le 4.5\%)$ indicate that the position of the minimum is fairly insensitive with respect to large variations of x. This may be expected because replacement of Ce by La in CeAl₂ will not lead to appreciable variations of $n(E_F)$.

Alternative explanations of the resistance anomaly in $CeAl_2$ might be sought in the presence of critical fluctuations of the magnetization in the vicinity of the Néel temperature^{13,14} or of complex magnetic ordering.¹⁵ Within the framework of existing theory an appreciable influence of the former effect seems improbable because $k_{\rm F}d$, the product of Fermi wave vector of the conduction electrons and nearest-neighbor distance of the Ce ions, has a large value (≈ 5). Moreover, an enlargement of d as effectuated by partial replacement in CeAl₂ of Ce by La does not alter appreciably the shape of the resistance anomaly.

Complex ordering, if present at all, does not seem to have an influence because in the system $Ce_{1-x}La_xAl_2$ with increasing value of x, the resistance minimum remains at about the same temperature whereas the ordering temperature is drastically reduced. It may be concluded that critical fluctuations of the magnetization or complex magnetic ordering do not seem to be effective in CeAl₂.

Thanks are due to Dr. Maranzana and Dr. Havinga for fruitful discussions and to P. van Aken for his assistance in the measurements.

³T. Sugawara and S. Yoshida, J. Phys. Soc. Japan <u>24</u>, 1399 (1968).

⁴A. S. Edelstein, Phys. Letters <u>27A</u>, 614 (1968).

 $^5\mathrm{H.}$ H. Hill, W. N. Miner, and R. O. Elliott, Phys. Letters 28A, 588 (1969).

⁶R. O. Elliot, H. H. Hill, and W. N. Miner, Phys. Status Solidi 32, 609 (1969).

⁷M. B. Maple and Z. Fisk, in <u>Proceedings of the</u> Eleventh International Conference on Low Tempera-

ture Physics, St. Andrews, Scotland, 1968, edited by

J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968), Vol. II, p. 1288.

⁸H. J. van Daal and K. H. J. Buschow, Solid State Commun. 7, 217 (1969).

⁹A. S. Edelstein, Phys. Rev. Letters <u>20</u>, 1348 (1968).

¹⁰J. Kondo, Progr. Theoret. Phys. (Kyoto) <u>32</u>, 37 (1964), and <u>34</u>, 523 (1965).

¹¹Y. Nagaoka, Phys. Rev. <u>138</u>, A1112 (1965).

¹²M. P. Sarachik and G. S. Knapp, J. Appl. Phys. <u>40</u>, 1105 (1969).

¹³P. G. de Gennes and J. Friedel, J. Phys. Chem. Solids 4, 71 (1958).

¹⁴M. E. Fisher and J. S. Langer, Phys. Rev. Letters <u>20</u>, 665 (1968).

 $\overline{}^{15}$ R. J. Elliott and F. A. Wedgwood, Proc. Phys. Soc. (London) <u>81</u>, 846 (1963).

¹T. Sugawara, J. Phys. Soc. Japan 20, 2252 (1965).

²T. Sugawara and H. Eguchi, J. Phys. Soc. Japan <u>21</u>, 725 (1966).