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MAGNETIC ORDERING IN $\text{Ce}_3\text{Al}_{11}$ (*)

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Résumé. — Les propriétés magnétiques de $\text{Ce}_3\text{Al}_{11}$ ont été étudiées sous pression à basses températures. Il semble que certains des effets observés dans CeAl_3 soient dus à la présence en faible quantité d'une phase parasite $\text{Ce}_3\text{Al}_{11}$.

Abstract. — The magnetic properties of $\text{Ce}_3\text{Al}_{11}$ have been studied at low temperatures under pressure. We show that some of the striking properties of CeAl_3 may be due to the presence of small amounts of $\text{Ce}_3\text{Al}_{11}$.

Among the Ce compounds, those with Al exhibit striking properties : Kondo behaviour has been observed in CeAl_2 , CeAl_3 and probably in $\text{Ce}_3\text{Al}_{11}$ at high temperatures [1, 2, 3, 4].

However the low temperature properties of these three compounds are very different. CeAl_2 is ordered at $T = 0$ but for CeAl_3 the situation is not clear. Its low temperature properties seem to be characteristic of a non-magnetic ground state : high linear temperature term of the specific heat, T^2 dependence of the resistivity [5]. CeAl_3 could be a Kondo lattice at $T = 0$. However some other properties namely the presence of two well marked peaks in the specific heat at 2.5 and 6 K [2] are unexplained. We have recently shown that a weak remanent magnetization appears just at 6 K and increases down to 2.5 K where it decreases abruptly to a very low value. Moreover under a 6 kbar pressure only the anomaly at 6 K remains in the specific heat; this phenomena is correlated to the persistence of the remanent magnetization below 2.5 K. If these properties are intrinsic to CeAl_3 , this compound could be the first example of a compound magnetically ordered between two temperatures and finally non-magnetic at low temperature [6]. However the purity of CeAl_3 is a crucial problem as this

compound does not exist in the liquid phase. We have shown [2] that the peak observed previously at 4.2 K in the specific heat was due to the presence of a parasitic phase of CeAl_2 and could be eliminated by an appropriate annealing.

In this same spirit, we have investigated the magnetic properties of $\text{Ce}_3\text{Al}_{11}$ in order to check whether or not the observed properties of CeAl_3 , namely the two peaks at 6 K and 2.5 K in the specific heat are due to the presence of a parasitic phase.

The polycrystalline sample was prepared in an a.c. furnace by a semi-levitation method.

The magnetization was measured at the S.N.C.I. Laboratory in Grenoble 1) between 1.4 and 4.2 K in a maximum field of 15 T by a classical extraction model, 2) between 4 K and 300 K in a vibrating pick-up coil experiment. a.c. susceptibility measurements have been also performed below 15 K down to 1.2 K. The two series of measurements were performed at $P = 0$ and $P \simeq 6$ bars in a CuBe sample holder.

1. Experimental results. — 1) Above 9 K the susceptibility follows a Curie-Weiss law at least up to 100 K with a paramagnetic Curie temperature $\theta \simeq -25$ K and an effective moment μ_{eff} determined without any correction, equal to $2.2 \mu_B$ in good agreement with previous results [3]. Both θ and μ_{eff} are pressure independent.

(*) This paper has been presented at the LT 15 Conference as a post-deadline paper.

2) Below 6 K a more complicated behaviour is observed. The compound exhibits a remanent magnetization which increases up to a maximum value of $0.1 \mu_B/\text{at. Ce}$ at 4.2 K, decreases abruptly at 2.5 K and then remains constant at lower temperatures (Fig. 1), with a very low value of $\approx 0.01 \mu_B/\text{at. Ce}$. Between 2.5 and 6 K (Fig. 2) the magnetization curves show a ferro-magnetic-type behaviour, however with an unusual

FIG. 1. — Remanent magnetization of $\text{Ce}_3\text{Al}_{11}$ at $P = 0$ and $P = 6$ kbars between 1.4 and 4.2 K.

FIG. 2. — Magnetization curves of $\text{Ce}_3\text{Al}_{11}$ between 0 and 5 kOe at $P = 0$ in the range 1.4–4.2 K.

shape which may be due to the strong anisotropy of the orthorhombic lattice [7]. The magnetization, which is twice its remanent value at 1 kOe, reaches only $0.7 \mu_B/\text{at. Ce}$ in 150 kOe at 1.4 K (Fig. 3). A simple picture is that in a first step the magnetic moment is aligned, along each preferential axis of a microcrystal and that in a second step each component of the microcrystal is slowly oriented along the H magnetic field since the preferential axes are randomly distributed with respect to H .

In the temperature range corresponding to the low remanent magnetization ($T < 2.5$ K) a transition occurs in the magnetization curves. It is similar to a metamagnetic transition but is not first order. The slope dM/dH is always much lower than the reciprocal demagnetization factor. Above the critical field the magnetization reaches the same value as in the ferromagnetic regime.

FIG. 3. — Magnetization of $\text{Ce}_3\text{Al}_{11}$ in 150 kOe at 4.2 and 1.43 K (upper curves). The magnetization curve of a sample of CeAl_3 at 4.2 K is given for comparison (lower curve).

3) Under a pressure of 6 kbars the *abnormal* behaviour no longer exists below 2.5 K. As shown on the figure 1 the remanent magnetization which continues to appear abruptly at 6 K has only a slight decrease below 2.5 K.

4) Recently, by measuring different samples of CeAl_3 , we have concluded that the remanent properties, previously detected between 2.5 and 6 K [2, 6] are due to the presence of a parasitic phase of $\text{Ce}_3\text{Al}_{11}$. For different samples, the $\text{Ce}_3\text{Al}_{11}$ content varies from ten to one percent. For one sample, no evidence of the $\text{Ce}_3\text{Al}_{11}$ phase has been observed whereas that of CeAl_2 has been clearly detected by the presence of only one weak specific heat anomaly at the ordering temperature of CeAl_2 (3.8 K). The intrinsic specific heat of CeAl_3 is presently unknown. The high sensitivity of the samples not only to the parasitic phases but also to the defects [8] leads to a quite difficult problem. The experimental situation is similar to that known for other instable valency compounds such as TmSe [9]. Despite this difficulty, the difference between CeAl_3 and the two well-ordered compound CeAl_2 and CeIn_3 is clearly shown by the temperature dependence of the entropy, since this representation masks the weak parasitic phase anomaly. For CeAl_2 and CeIn_3 a marked decrease of the entropy is observed at the temperature of the maxima of the specific heat whereas for CeAl_3 the entropy follows a smooth temperature dependence.

2. Conclusion. — The two peaks generally observed in the specific heat of CeAl_3 at 2.5 and 6 K seem due to the $\text{Ce}_3\text{Al}_{11}$ parasitic phase, they have been unambiguously related to the remanent properties.

Few experiments have been made on $\text{Ce}_3\text{Al}_{11}$ until now. A better understanding of its low temperature properties is crucial for the interpretation of the CeAl_3 behaviour. $\text{Ce}_3\text{Al}_{11}$ is a striking case : the spectacular disappearance of one anomaly under pressure whereas the other anomaly is pressure independent is unex-

plained. An interesting problem is the corresponding variation of the specific heat notably its low temperature behaviour. A future comparison with the two

other magnetically well ordered compounds CeIn_3 and CeAl_2 will be fruitful for the understanding of these abnormal compounds.

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