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MAGNETIC PROPERTIES OF THE Ce-Rh BINARY PHASES

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Abstract. – Crystallographic, magnetic and resistivity studies on the Ce-Rh binary phases clearly define two Ce ground state regions: i) $CeRh_3$, $CeRh_2$ and CeRh as intermediate valence compounds and ii) Ce_5Rh_4, Ce_3Rh_2 Ce_5Rh_3 and Ce_7Rh_3 with magnetic transitions at low temperature.

Some Ce-Rh compounds, such as $CeRh_2$ [1] and $CeRh_3$ [2] are recognized as typical examples of intermediate valence (IV) systems with a nearly temperature independent magnetic susceptibility (χ_0) and a strong reduction of their volume (ΔV) with respect to the isomorphous trivalent rare earth compounds. The existence of other compounds in the Ce-Rh system, like CeRh, Ce_5Rh_4, Ce_5Rh_3, Ce_3Rh_2 and Ce_7Rh_3, was reported concerning their structure [3] and their high temperature susceptibility [4, 5].

In order to improve the knowledge on this binary system, a systematic study of these compounds was carried on their crystallographic structure and ground state properties.

Seven binary phases were recognized in this system. Their structure-type, mean atomic volume (V_{at}), magnetic transition temperature (T_N or T_c), saturation magnetization (M_s) or susceptibility (χ_0), extrapolated Curie-Weiss temperature θ_P or temperature of the maximum of the susceptibility (T_M) are given in the table I.

The crystalline structure and the lattice parameters of the different compounds are in agreement with reference [3]. This study confirms that the Ce_4Rh_3 and Ce_3Rh compounds do not exist. Concerning the magnetic properties different behaviours are clearly shown between the Rh rich region and the Ce rich one. All the intermediate valence compounds belong to the Rh rich side and their magnetic susceptibility are weakly temperature dependent, with nevertheless an increase at low temperature. The origin of these upturns has been analysed in the cases of $CeRh_3$ and CeRh (the results of $CeRh_2$ are taken from Ref. [7]). For $CeRh_3$, the susceptibility is independent of the magnetic field (H $\leq$ 15 Tesla), indicating a possible intrinsic origin of the low temperature increase of $\chi(T)$. In the case of CeRh the $\chi(T)$ variation, for $T \leq 200$ K, is well described by the relationship $\chi(T) = \chi_0 + C_{imp} / T + aT^2$ with $C_{imp} = 5 \times 10^{-3}$ emuK/mole, $a = 1.8 \times 10^{-8}$ emu/K^{2}mole and $\chi_0 = 0.52 \times 10^{-3}$ emu/mole. This leads to assume that, in CeRh, the low temperature upturn is essentially due to impurity contributions.

Table I. – *Structure, mean atomic volume* (V_{at}), *susceptibility* (χ_0, *corresponding to* χ (4.2 K) *for* $CeRh_3$ *and* $CeRh_2$), *saturation magnetization* (M_s) *at* 4.2 K, *magnetic transition temperature* (T_c *or* T_N) *and the extrapolated Curie-Weiss temperature* θ_p *or the temperature of the maximum of* $\chi(T)$ T_M, *in the* Ce-Rh *system. The* M_s *values are extracted from* $M(H,$ 4.2 K) *up to* $H = 17.5$ Tesla. $a = 10^{-3}$ emu/mole.

Compound	Structure type	V_{at} (Å^3)	χ_0 (a)	M_s (μ_B/Ce)	T_N, T_c^* (K)	T_M, θ_P^* (K)
$CeRh_3$	$AuCu_3$	16.2	0.54	–	–	> 800
$CeRh_2$	$MgCu_2$	17.8	1.30 [7]	–	–	380 [7]
CeRh	CrB	22.0	0.52	–	–	270
Ce_5Rh_4	Sm_5Ge_4	23.3	–	0.32	0.8	-200^*
Ce_3Rh_2	Er_3Ni_2	24.7	–	0.74	3.6	-37^*
Ce_5Rh_3	Pu_5Rh_3	25.0	–	0.70	4.4/2.5	-45^*
Ce_7Rh_3	Th_7Fe_3	27.7	–	0.69	7.2^*	13^* [4]

The electrical resistivity of CeRh shows a good metallic behaviour at $T \to 0$ K, with $\rho_0 = 1.1$ $\mu\Omega$cm and a significant increase up to 65 $\mu\Omega$cm at 300 K. As it is typically observed in Ce IV compounds, T_M ($T_M \simeq 270$ K) is in coincidence with the temperature where the slope of $\rho(T)$ becomes softer.

On the Ce rich side, all the compounds show magnetic order at low temperature. The figure 2 illustrates the thermal variation (for $T \leq 10$ K) of the initial susceptibility $\chi(T)$ of the antiferromagnetic Ce_5Rh_4 Ce_3Rh_2 and Ce_5Rh_3 and the low field magnetization

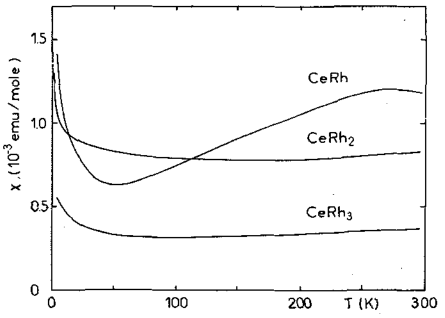

Fig. 1. Thermal variation of the susceptibility of CeRh, $CeRh_2$ [7] and $CeRh_3$ for $4 \leq T \leq 300$ T.

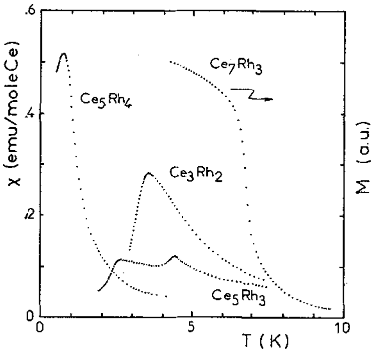

Fig. 2. – Low temperature magnetic behaviour of Ce_7Rh_3, Ce_3Rh_2, Ce_5Rh_3 and Ce_5Rh_4 for $T \leq 10$ K.

$M(T,\ H = 1$ mT) of the ferromagnetic Ce_7Rh_3 compounds.

The temperatures T_M of the IV compounds and θ_P of the magnetic ground state systems, which follow a monotonic dependence with the relative concentration, are related with the stability of the Ce magnetic moment.

A particular attention was given to Ce_5Rh_4 because it represents the limit for the Ce trivalency with respect to VI CeRh, with only 5 at % of difference in their relative volume. For $T > 100$ K, the $\chi(T)$ variation follows a Curie-Weiss law with a Curie constant $C = 0.802$ emuK/mole Ce and a large $|\theta_P| = 200$ K. At lower temperature the $\chi(T)$ dependence increases faster than the Curie-Weiss law, this behaviour cannot be understood only in terms of crystal field effects.

In the Ce_5Rh_4 elementary cell, there are three positions for the Ce atoms, with respective pointset at 4c and two equivalents at 8d. From the entropy gain ($0.34R \ln 2$, $T \leq 10$ K) and the low saturation magnetization (0.32 μ_B/Ce), we assume that only one third of the Ce atoms (those in the 4c positions) are involved at the $T_N = 0.8$ K magnetic transition. This is confirmed by the tentative analysis of the low temperature susceptibility ($T \leq 100$ K) in two terms: $\chi(T) = C/T + \chi_0$, with $C = 0.095$ emuK/mole Ce and $\chi_0 = 2 \times 10^{-3}$ emu/mole Ce. Therefore the Ce ground state in Ce_5Rh_4 could be a mixture of magnetic and Kondo-like states with completely different characteristic temperatures.

The $\chi(T)$ dependence of Ce_5Rh_3 (Fig. 2) shows two magnetic transitions which are field independent up to 0.5 T. These transitions are clearly away from those of the other magnetic phases of the Ce-Rh system and are therefore characteristic of the magnetic structure of this compound, with three different sites for Ce.

The only compound which orders ferromagnetically is Ce_7Rh_3, with $T_c = 7.2$ K (Fig. 2). This is also the only compound where the atomic volume can be extracted from a Végard's law between Rh and γ-Ce (Fig. 3).

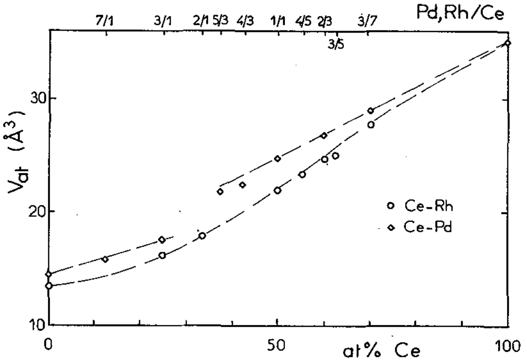

Fig. 3. – Mean atomic volume (V_{at}) as a function of the relative concentration in the Ce-Pd (◇) and Ce-Rh (○) systems.

It is very instructive to compare the Ce-Rh binary system with that of Ce-Pd. In figure 3, the atomic volume V_{at} is plotted as a function of the Ce concentration for both binary systems. In the Ce-Rh system the change of V_{at}, with the relative concentration follows a continuous curve, including the region between the IV and magnetic compounds, where the Ce-Pd system does show a discontinuity [6]. Only four compounds (on seven studied in each system) have the same crystalline structure and only two pairs show similar magnetic behaviour, i.e. $CeRh_3$ and $CePd_3$ as IV compounds and Ce_3Rh_2 and Ce_3Pd_2 as antiferromagnets.

Another striking feature in this comparaison is that Ce_7Rh_3 orders ferromagnetically, while the isostructural Ce_7Pd_3 presents an antiferromagnetic state [6].

Finally, there is a very important difference between the Ce valence in the isostructural CeRh and CePd. Such a difference in the Ce valence state cannot be due to the weak difference between the atomic volume of the partners, but mainly to their electronic difference.

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