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To cite this version:

J. Daou, P. Vajda, G. Hilscher, N. Pillmayr. MAGNETIC AND THERMAL PROPERTIES OF Tm AND $\alpha$ - TmHx. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-357-C8-358. <10.1051/jphyscol:19888160>. <jpa-00228308>

HAL Id: jpa-00228308
https://hal.archives-ouvertes.fr/jpa-00228308
Submitted on 1 Jan 1988

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MAGNETIC AND THERMAL PROPERTIES OF Tm AND $\alpha$ – TmH$_x$

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Abstract. – We present specific heat measurements of Tm (99.99 %) and $\alpha$ – TmH$_x$ as well as resistivity measurements on single crystals of the same material. The $c$-axis resistivity results can be explained in terms of a hydrogen dependent evolution of superzone boundaries while the $b$-axis data and the magnetic heat capacity cannot simply be analysed with a ferrimagnetic contribution, but indicate a complex magnetic structure below 4.2 K which becomes more pronounced as the hydrogen content increases.

Tm possesses a large solid solubility of H down to low temperatures in the $\alpha$-phase ($\alpha$ – TmH$_x$, $x_{\text{max}} = 0.11$ [1]) which provides an excellent opportunity to study the effect of hydrogen upon its magnetic and electronic properties. Thus recent investigations of resistivity and magnetisation of polycrystalline specimens revealed that both $T_N$ (57.5 K) and $T_C$ (39.5 K) decrease with rising H – content down to 45.5 K and 29 K respectively for $x = 0.1$. The results were explained in terms of a depopulation of the metallic d band, decreasing the number of conduction electrons responsible for the RKKY interaction, and by the formation of a H – H pair configuration below 170 K modifying the magnetoelastic interaction [1]. In order to contribute further to a clarification we performed specific heat measurements on carefully outgassed bulk 99.99 % Tm, prepared by Ames Labs., on samples of about 1.5-2 g subsequently hydrogenated at 500 °C up to $x = 0.1$.

In figure 1 we present the specific heat in the vicinity of $T_N$ (insert) and the low temperature data in an $(C_p - C_N)/T$ versus $T^2$ plot with the nuclear heat capacity $C_N = 28 T^{-2}$ mJ/mole.K. These data were analysed with a standard least square routine (NAGLIB,E04KCF). However, the magnetic contribution to the heat capacity, $C_m$, could not be determined unambiguously, in particular for samples with higher $x$ values. According to the magnetic structure which is sinusoidally modulated antiferromagnetic below $T_N$ and gradually squares up to yield an antiphase ferrimagnet below $T_e$ = 39 K, with three spins up and four spins down, we expect for $C_m$ with a non-negligible anisotropy: $C_m = \mu T^{5/2} \exp(-\Delta/kT)$. This term could indeed be detected using for $C_p = \gamma T + \beta T^3 + C_N + AT^{-2}$, where the last term represents $C_N$ mentioned above and $\gamma$ and $\beta$ the coefficients of the electronic and lattice contributions to $C_p$. In the wide temperature range from 1.5 to 15 K we derived for pure Tm in units of mJ/mole.K$^n$: $\gamma = 10.5$, $\beta = 0.73$, $\mu = 554$, $A = 28$ and $\Delta/k = 43$ K. For TmH$_{0.03}$ we observed an increase of $\gamma$ up to 17.5, but a dramatic decrease of $\mu$ and $\Delta$ of nearly one order of magnitude, the agreement between the fit and experiment, however, was hardly satisfying.

In this connection it is worthwhile to note that widely conflicting results (except for $C_N$) have been published for pure Tm, whereby different power laws have been used to describe $C_m$ [2-4]. A comparison of those results with our data shows clearly that the former are much higher and fall into the range between the graphs of $\alpha$ – TmH$_x$, 0.03 < $x$ < 0.06 in figure 1, with a typically analogous temperature dependence as for $\alpha$ – TmH$_x$. This demonstrates that the $C_p$ measurements of "pure" Tm published and known to us can be referred to as those of $\alpha$ – TmH$_x$ with an appreciable content of H.

The $C_p$ measurements of $\alpha$ – TmH$_x$ with larger $x$ values ($x > 0.03$) could neither be fitted acceptably with the expression given above nor with a modified rela-
tion for $C_m$ using various power laws with and without an anisotropy gap. This led us to further resistivity and magnetisation measurements on single crystals. Preliminary magnetisation measurements performed in collaboration with Chouteau (SNCI Grenoble) indicate that below 4.2 K a not yet resolved complex magnetic structure occurs which seems to become more pronounced the H content increases.

The electrical resistivity measurements were performed parallel to the c- and b-axis on two single crystals of Tm loaded successively with H up to $x = 0.103$. For the pure metal, they give results in good agreement with the original data by Edwards and Legvold [5]. In figures 2 and 3 we present a global view of a selected set of measurements on $\alpha-TmH_x$ crystals parallel to b and c, respectively, showing a striking difference in the evolution of the two crystal orientations with $x$. While the b-axis crystals exhibit almost a linear increase of $\rho_b$ with $x$ (Fig. 2) giving an added H-resistivity contribution of $\rho_H^b = 4.0 \pm 0.2 \mu\Omega\,cm/\% \, H$, the apparent increase of $\rho_c^c$ is much larger and nonlinear. In fact the resistivity decrease due to the ferromagnetic ordering below 40 K is strongly suppressed by hydrogen in solution, completely disappearing for $x > 0.05$ (Fig. 3). We attribute this to the evolution of the magnetic superzones caused by hydrogen, a phenomenon which had already been observed by us to a smaller degree in $\alpha$-ErH$_x$ single crystals [6]. Following reference [5] who have fitted the Tm data using the Elliot-Wedgwood theory such as $(d\rho^b/dT)/(d\rho^c/dT) = (\rho^b_0/\rho^c_0)(1 - \delta M)^{-1}$ where the slopes are taken at room temperature, we find a superzone gap $\delta M$ of 0.73 for pure Tm (compared to the value of 0.86 from Ref. [5]) decreasing gradually to $\delta M = 0.4$ for TmH$_{0.1}$. A further direct result of figures 2 and 3 is the decrease of $T_N$ in both types of crystals with $x$, quite linearly by 1 K/at % H, confirming the polycrystal data and thus the purely electronic mechanism for its variation. In the same way, the spin disorder resistivity, deduced from figure 2 for the b-crystals, decreases linearly from $\rho_{b,0}^b = 25 \mu\Omega\,cm$ for pure Tm to 165 $\mu\Omega\,cm$ for TmH$_{0.1}$. The insert of figure 2 shows as an interesting phenomenon a strong increase of the magnetic contribution $\rho_{mag}(T)$ with $x$ beginning at the lowest temperatures. We have tried to fit $\rho_{mag}(T)$, both with power functions and with an exponential expression for the anisotropy gap $\Delta$. Similarly to the $C_m$ analysis we find a reasonable exponential fit over nearly two decades in the case of pure Tm (for both the b- and c-crystal orientations in the temperature range from 8 to 25 K). For $x > 0$, however, one has to assume for $\rho_{mag}^b(T)$ a sum of two contributions, $\rho_{mag}^b(T) = AT^n + BT^2 \exp(-\Delta/kT)$, with both $n$ and $\Delta$ decreasing with rising $x$.

In conclusion, we state that the $C_p$ measurements of "pure" Tm published up to now can be regarded as those of $\alpha-TmH_x$ and suppose from $\rho(T)$ and $C_p(T)$ measurements that an anisotropy gap occurs in the spin wave spectrum which is reduced with rising H-content going hand in hand with the evolution of a complex magnetic structure below 4 K.