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Low temperature susceptibility of LaNi_5 hydrides

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Résumé. — La susceptibilité paramagnétique à 4,2 K de LaNi_5 hydrogéné décroît avec la quantité d'hydrogène absorbé. Nos mesures montrent que la formation de nickel libre se produit lors de la désorption et non lors de l'absorption d'hydrogène.

Abstract. — The 4.2 K paramagnetic susceptibility of LaNi_5 hydrides decreases with the hydrogen content. Our measurements show that the formation of nickel particles occurs at the desorption and not at the absorption of hydrogen.

Among the intermetallic compounds, LaNi_5 is one of the most interesting because of the very large amount of hydrogen it can absorb. The magnetic properties of the various compounds vary in different ways upon hydriding. Whatever the mechanism of hydrogen absorption can be, for some of them such as $\text{RFe}_2[\text{I}]$ ($\text{R} = \text{Ce}, \text{Y}, \text{Lu}, \text{Sc}$) the hydriding increases the magnetic moment, while in others such as compounds with Co the moment is decreased. Therefore it is of importance to try to understand which mechanism causes the change in magnetic properties after hydrogen absorption.

We have studied the magnetization of the compound LaNi_5 at low temperature, in high fields as a function of the amount of absorbed hydrogen. The samples were prepared by fusion in cold levitation crucibles. The starting materials from Johnson-Matthey are 99.99 % wt. purity for La and 99.999 wt. % for Ni. To improve homogeneity the samples were repeatedly remelted (three times) and were annealed for 1 day at 1 300 °C and cooled to room temperature by quenching on the cold crucible. All the preparations and the annealings were made under purified argon atmosphere. The metallographic examination showed no other phase and revealed a large grain structure of the compound.

The X-ray diffraction data were obtained on a Phillips X-ray powder diffractometer. The X-ray

diagram firstly confirmed the non-existence of a second phase and gives for the lattice constants :

$$a = 5.017 \pm 0.003 \text{ \AA}$$

$$c = 3.983 \pm 0.003 \text{ \AA}.$$

For magnetic measurements the sample was put in a high purity copper (99.999 %) container, sealed under vacuum. Charging with hydrogen is achieved without putting the specimen in contact with atmospheric air. Thus, contamination by water or oxygen is very small. Furthermore the contamination due to the oxygen present in the equivalent dead volume is completely negligible. The quantity of absorbed hydrogen is determined by weighing the sample. The sample mass is of the order of 1.2 g. In the figure 1

Fig. 1. — Magnetization curves of the fresh (1) and fully hydrided (2) sample.

we have shown the magnetization curves up to 11.7 T at 4.2 K obtained after various charging with hydrogen.

Curve (1) corresponds to pure LaNi_5 without hydrogen. No curvature can be observed indicating the absence of particles of pure nickel. We find for the susceptibility :

$$\chi = 5.25 \times 10^{-6} \text{ emu/g}$$

in good agreement with the recent results of the literature [2]. The complete charge we obtain corresponding to the formula $\text{LaNi}_5\text{H}_{6.9}$ gives the curve (2). This shows : i) The magnetization M is no more proportional to the field H . An extrapolation of the high field linear part of $M(H)$ to $H = 0$ gives an intercept M_s corresponding to the saturation magnetization of a magnetic phase. The thermal variation of M_s suggests that it is due to pure nickel particles. A comparison of M_s to the saturation moment of nickel allows us to evaluate the amount of pure nickel present in the sample. We calculate 0.3 mg (i.e. $\approx 0.02\%$ weight of the sample). ii) Considerable decrease in the susceptibility :

$$\chi = 1.05 \times 10^{-6} \text{ emu/g}.$$

The same behaviour has been observed recently but with a smaller decrease probably due to a smaller hydrogen content [3].

A second set of measurements was performed in the following way :

The sample was fully desorbed and then gradually recharged in pure hydrogen. The quantity of hydrogen absorbed is increased at room temperature by simply increasing the pressure of hydrogen gas, without desorbing the sample. The complete procedure is finally the following : a) Pure LaNi_5 ; b) Full hydriding; c) Full dehydriding; d) Successive partial hydridings up to full hydrogen absorption.

Fig. 2. — Magnetization curves of LaNi_5 after complete desorption and partial hydriding : (3) $\text{H/LaNi}_5 = 0$; (4) $\text{H/LaNi}_5 = 0.6$; (5) $\text{H/LaNi}_5 = 1.85$; (6) $\text{H/LaNi}_5 = 2.68$; (7) $\text{H/LaNi}_5 = 5.25$.

The results are shown on figure 2 : After complete dehydriding, the magnetization curve is no longer linear (curve (3)) as in the initial state. A rather large amount of pure nickel, 1.3 mg, has been formed and furthermore the high field susceptibility is smaller

than in the fresh sample : $\chi = 4.74 \times 10^{-6} \text{ emu/g}$. However the possibility of formation of H_2 traps cannot be completely excluded.

Curves (4) to (7) on figure 2 show the magnetization *versus* field upon increasing hydriding. No further formation of nickel particles is shown and the susceptibility decreases continuously. As shown on figure 3,

Fig. 3. — Variation of the susceptibility with the ratio H/LaNi_5 .

the linear part of the curve (H/LaNi_5) extrapolates to zero at $\text{H/LaNi}_5 \approx 8.5$ which is very near the maxima value $\text{H/LaNi}_5 = 9$ deduced from structural considerations. The results are summarized in the table I.

Table I.

H/LaNi_5	$\chi (10^{-6} \text{ emu/g})$	Curve
0	5.25	1
6.9	1.05	2
0 (^a)	4.74	3
0.60	4.0	4
1.85	3.43	5
2.68	2.91	6
5.25	1.77	7

(^a) : After complete desorption.

Our results show that the formation of pure nickel particles is not a consequence of the hydrogen absorption but rather of the desorption. Indeed, the saturation magnetization is strongly increased by complete dehydriding and does not change upon hydriding. Therefore it is difficult to understand how the mechanism proposed in ref. [4] can work. It must be noticed that generally the different authors observe an increase of the amount of pure nickel particles after several cycles. In no way the experimental procedure allow them to conclude whether the formation of these particles occurs during the hydriding or the dehydriding.

The decrease of χ with the hydrogen content as

shown in figure 3 can be interpreted as a decrease of the density of states at the Fermi level. This implies, as shown by EPR measurement in hydrided LaNi_5 , doped with Gd [5], a charge transfer from H to Ni or an equivalent mechanism.

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