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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 RF$_2$ [1] (R = Ce, Y, Lu, 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-Mattey 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 1300 °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:

\[
\begin{align*}
a &= 5.017 \pm 0.003 \text{ Å} \\
c &= 3.983 \pm 0.003 \text{ Å}
\end{align*}
\]

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

![Magnetization curves of the fresh (1) and fully hydrided (2) sample.](image)
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 LaNi5 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 cor-
responding to the formula LaNi5H6.9 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) Consi-
derable 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 LaNi5 ; b) Full hydri-
ding ; c) Full dehydriding ; d) Successive partial
hydridings up to full hydrogen absorption.

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 \( \text{H}_2 \) 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 suscepti-
bility decreases continuously. As shown on figure 3,

\[ \chi/10^{-6} \text{ emu/g} \]

Fig. 2. — Magnetization curves of LaNi5 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) \( \text{H/LaNi}_5 = 5.25 \).

\[ \chi/10^{-6} \text{ emu/g} \]

Fig. 3. — Variation of the susceptibility with the ratio \( \text{H/LaNi}_5 \).

\[ \chi/10^{-6} \text{ emu/g} \]

Table I.

<table>
<thead>
<tr>
<th>( \text{H/LaNi}_5 )</th>
<th>( \chi/10^{-6} \text{ emu/g} )</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 (*)</td>
<td>4.74</td>
<td>3</td>
</tr>
<tr>
<td>0.60</td>
<td>4.0</td>
<td>4</td>
</tr>
<tr>
<td>1.85</td>
<td>3.43</td>
<td>5</td>
</tr>
<tr>
<td>2.68</td>
<td>2.91</td>
<td>6</td>
</tr>
<tr>
<td>5.25</td>
<td>1.77</td>
<td>7</td>
</tr>
</tbody>
</table>

(*) : After complete desorption.

Our results show that the formation of pure nickel
particles is not a consequence of the hydrogen absorp-
tion 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 experi-
mental procedure allow them to conclude whether
the formation of these particles occurs during the
hydriding or the dehydriding.

The decrease of \( \chi \) 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 LaNi₅, doped with Gd [5], a charge transfer from H to Ni or an equivalent mechanism.

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References