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MAGNETIC SUSCEPTIBILITY OF NICKEL HYDRIDE

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Abstract. - The magnetic susceptibility of a series of nickel hydride samples has been measured in magnetic fields 0.9-9 kOe and in the temperature range 1.4-100 K. The susceptibility is paramagnetic and yields an estimate of $\chi = 1.2 \times 10^{-6}$ emu g⁻¹ as an upper limit for the ideal nickel hydride matrix.

Introduction

The magnetic properties of many transition metals undergo drastic changes when hydrogen is interstitially absorbed into the metal matrix. Pure palladium has an exchange enhanced paramagnetic susceptibility. When increasing amounts of hydrogen are introduced into the metal the susceptibility is decreased. Palladium hydride becomes diamagnetic and even superconducting [1]. Chromium is a body centered cubic antiferromagnet. The hexagonal chromium hydride has metallic properties and a strongly enhanced paramagnetic susceptibility. These properties are in many respects similar to those of palladium [2]. Absorption of interstitial hydrogen into the face centered cubic nickel lattice destroys the ferromagnetic properties of nickel and the magnetic susceptibility becomes paramagnetic [3]. In these samples, having a hydrogen to nickel ratio ≥ 0.7 , ferromagnetic impurities of the order of 100 ppm of iron yielded a contribution to the paramagnetism. In order to find out whether the ideal pure nickel hydride matrix really is paramagnetic, or possibly diamagnetic, we were motivated to measure the susceptibility, its field and temperature dependence, of a series of nickel hydride samples. The susceptibility samples need to have a large enough mass in a small volume. In order to obtain a complete hydrogenation, with all the octahedral interstitial sites of the nickel lattice filled with hydrogen, the samples were prepared by application of a high pressure hydrogen atmosphere [4]. This is in contrast to the earlier investigation in which the hydrides were electrochemically formed.

Materials and methods

The nickel hydride samples were prepared from Ni wire of 99.999 % purity, that was successively rolled down to foils of 10 or 15 μm thickness. The samples were charged in a high pressure (~ 1.5 GPa) hydrogen atmosphere. The hydrogen to nickel ratio, about 1:1, was determined by mass spectroscopy analysis. In order to prevent decomposition the samples were kept in liquid nitrogen after hydrogenation. Samples 2, 3 and 4 were checked for ferromagnetism in an ac magnetometer [5]. With this a magnetization of the order of 1 % of the total nickel content may be detected. Only samples with a residual ferromagnetism below this limit were accepted for susceptibility measurements. In order to reach complete hydrogenation, according to this criterion, sample 2, that was ob-

tained by rehydrogenating the foil of sample 1, had to be charged four times. The other samples were hydrogenated once. The susceptibility measurements were made with the Faraday method in magnetic fields from 0.9 kOe to 9 kOe and at temperatures from 1.4 K to 100 K [6]. After these measurements sample 4 was warmed stepwise to successively higher temperatures above 100 K, in the last cycle up to 280 K, whereafter its susceptibility was remeasured at 1.4 K, 4.2 K and 90 K.

Results and discussion

All the investigated nickel hydride samples exhibit a strong paramagnetic susceptibility. It is field dependent over the whole temperature range, 1.4 to 100 K. The susceptibility measured in a constant field is temperature dependent and increases monotonically as the temperature is lowered. This can be seen in figure 1. Here the susceptibility, χ , for the four samples measured in magnetic fields, H , of 0.9 kOe and/or 1.3 kOe is plotted *versus* temperature, T . The field dependence of the magnetization, $\chi \cdot H$, at constant temperature is shown in figure 2. In this one can see how

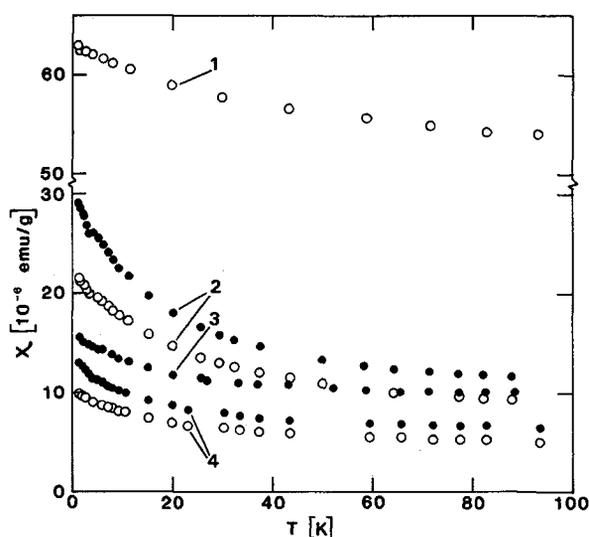


Fig. 1. - The magnetic susceptibility, χ , of four nickel hydride samples, numbered in the text, plotted *versus* temperature, T . The unfilled circles denote values measured in a magnetic field of 1.3 kOe and the filled 0.9 kOe.

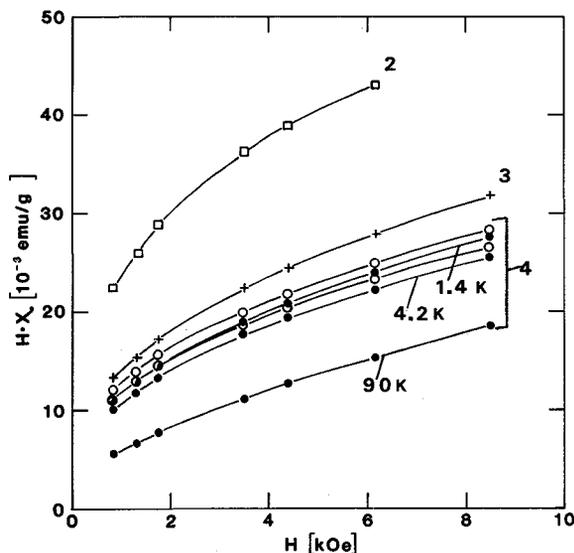


Fig. 2. - The magnetization $\chi \cdot H$ versus magnetic field, H , for three of the nickel hydride samples. Samples 2 and 3 were measured at 4.2 K. The filled circles denote sample 4 measured at 90 K, 4.2 K and 1.4 K. The unfilled circles show what happened to sample 4 when it was warmed repeatedly to about 200 K and remeasured at 4.2 K. The lines are drawn to guide the eye.

the field dependence of sample 4 develops as the temperature is decreased from 90 K to 4.2 K and 1.4 K. It is also shown how repeated warming of sample 4 to about 200 K, where it was kept for about 20 minutes, affects the magnetization at 4.2 K. We think that the difference between individual samples may be explained by their different history with regard to hydrogenation and temperature cycling. This can be seen not only in sample 4 as it is warmed, but also when the magnetic susceptibility of sample 1 is compared to that of sample 2. These were obtained from different hydrogenation cycles of the same foil. (Sample 1 was not included in figure 2 where it would be well outside the displayed range.) In the high pressure phase of nickel hydride there may be small scale local fluctuations in the hydrogen concentration, leaving some octahedral hydrogen sites unoccupied. By keeping the sample at liquid nitrogen temperatures this state can be frozen in non-equilibrium at atmospheric pressure. 200 K is well below the region where hydrogen desorption has been observed. The mobility of hydrogen is however sufficient to allow a rearrangement of hydrogen atoms in the lattice. Such phenomena have been suggested also in other magnetic studies [7]. Since the magnetic properties of the nickel atom depend crucially on the local environment, this rearrangement leads to a change in the susceptibility of the nickel hydride sample. It is evident from our results that there is a pronounced effect on the overall level of the susceptibility as well as on its field dependence. Since sample 4 has the lowest susceptibility and the smallest field dependence we use this to give an upper limit of the magnetic suscepti-

bility of the ideal nickel hydride matrix. If we assume that the main part of the field dependence at 90 K is due to the development of small islands of ferromagnetic nickel clusters we may use the relation

$$\chi = \chi_m + \sigma_s c / H \quad (1)$$

to yield the concentration, c , of ferromagnetic material with saturation magnetization, σ_s , in a matrix with susceptibility χ_m . In our limited field range we do not reach a complete saturation. Using the values of the susceptibility in 6.2 kOe and 8.5 kOe at 90 K for sample 4 we obtain $\chi_m = 1.2 \times 10^{-6} \text{ emu g}^{-1}$ and $\sigma_s \cdot c = 7 \times 10^{-3} \text{ emu g}^{-1}$. This corresponds to an amount of 1×10^{-4} of the total Ni content being ferromagnetic. This estimate is in good agreement with the results from the ac measurements of the residual magnetization.

In addition to the ferromagnetic Ni clusters there is also a contribution that may be attributed to impurities and/or smaller clusters of Ni. This is evidenced from the Curie-like increase of the susceptibility at low temperatures and from the fact that the magnetization does not saturate in fields below 9 kOe. We will analyse these data further and also make a more detailed study of the temperature dependence of the susceptibility after warming of the samples above the decomposition temperature. In this work we have shown that as the samples are more completely hydrogenated, the susceptibility decreases and we have given an estimate of the susceptibility for nickel hydride of $\chi = 1.2 \times 10^{-6} \text{ emu g}^{-1}$. This value might possibly be lowered if the metallurgical problems of preparing a completely hydrogenated sample for susceptibility measurements could be overcome.

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