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GIANT MOMENTS IN PdNi ALLOYS

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Résumé. — Nous avons étudié les effets des interactions dans les alliages PdNi et montré que les atomes isolés et les paires ne sont pas magnétiques. Les groupes de trois atomes sont magnétiques et induisent dans la matrice PdNi de larges moments. Ces résultats montrent que la transition vers l’état magnétique se fait dans les alliages PdNi de façon inhomogène.

Abstract. — We have studied the interaction effects in the PdNi alloys. We have shown that the isolated atoms and pairs are not magnetic. The groups of three Ni atoms are magnetic and are associated with a giant moment which is induced in the nearly magnetic PdNi host. Our results show that in the PdNi the transition toward the magnetic state is inhomogeneous.

In a number of concentrated Ni alloys [1,2] the transition from the non magnetic to the magnetic state is governed by the interactions which lead to the formation of giant moments. This paper deals with the study of the interaction effects and the occurrence of giant moments in the PdNi alloys.

We have measured the magnetization of PdNi and PdNi,Fe alloys between 0.05 and 4 K in fields \( h \leq 75 \) kOe. These samples were melted in alumina crucibles. The analysis shows that the actual concentration of Ni is very close to the nominal one but that small amounts of iron (\( \approx 50 \) ppm) were introduced during the melting. We have also measured the susceptibility of five other PdNi samples \((0 \leq y \leq 1.3\%\)\) between 4.5 and 300 K in a field \( h = 9.750 \) Oe using a Faraday balance. These samples were elaborated in an induction furnace by a semi-levitation method, in order to avoid the introduction of iron impurities during the melting.

The magnetization below 1 % is approximately proportional to the field (Fig. 1). Beyond 1 % a curvature appears which increases with the concentration. For \( h > 2 \) kOe, the magnetization in the range 1-2 % is independent of the temperature. For \( h < 2 \) kOe the magnetization at 1 K is smaller than at 0.1 K (Fig. 2). This effect is due to the magnetic impurities associated with giant moments since they are saturated in a field as low as 2 kOe at 1 K. For \( y < 1 \%\) after subtraction of the Palladium contribution \( \chi_{Pd} h \) and of the saturation magnetization \( \chi_{S} \) due to the magnetic impurities, the excess magnetization \( \Delta M \) can be written in the form:

\[
\Delta M(h) = M_1(h) y + M_2(h) y^2
\]

where \( M_1(h) \) is attributed to the isolated Ni atoms while \( M_2(h) \) shows the effects of the Ni-Ni interactions.
$M_2(h)$ exhibits a slight curvature beyond 50 kOe. The term $M_2(h)$ is not saturated even in fields as high as 75 kOe. This indicates that the impurities which contribute to $M_2(h)$ are not magnetic.

The excess susceptibility $\Delta \chi(T) = \chi_{\text{alloy}}(T) - \chi_{\text{Ni}}(T)$ measured at $h = 9750$ Oe between 4 and 300 K is represented, for $y < 1 \%$, by the law:

$$\Delta \chi(T) = \Delta \chi_1(T) y + \Delta \chi_2(T) y^2 .$$

The quantities $\Delta \chi_1(T)$ and $\Delta \chi_2(T)$, plotted in the diagrams $(1 + a\chi_{\text{Ni}})^2 / \Delta \chi_1$, $(1 + a\chi_{\text{Ni}})^2 / \Delta \chi_2$ [3, 4] as a function of the temperature, where $a$ is a parameter determined from the experimental data, exhibit Curie-Weiss behaviours above 50 K. (Fig. 4):

$$(1 + a\chi_{\text{Ni}})^2 / \Delta \chi_1 = (T - \theta_1) / C_1 ;$$

$$(1 + a\chi_{\text{Ni}})^2 / \Delta \chi_2 = (T - \theta_2) / C_2$$

with

$$\theta_1 = -25 \text{ K} , \quad \theta_2 = (-13 \pm 5) \text{ K} .$$

Within the local spin fluctuations theory [5] $\theta_1$ and $\theta_2$ can be interpreted as the fluctuation temperatures of the isolated atoms and of the pairs respectively. The value $\theta_1 = -25$ K is in agreement with the value deduced from specific heat measurements [6, 7]. $\theta_2$ is only two times smaller than $\theta_1$. This means that the Ni-Ni interactions are less effective than the Fe-Fe interactions in CuFe for example [8] where

$$\theta_1 = -29 \text{ K}$$

has approximately the same value as in PdNi but where the interactions are felt at several interatomic distances and $\theta_2$ for a pair of first neighbours would be very small.

We consider only the environment effects due to first neighbours.

Assuming that each Ni first neighbour of a Ni charges the Coulomb interaction $\Delta U = U_{\text{Ni}} - U_{\text{Pd}}$ by an amount $\delta U$, the fluctuation temperature $\theta_{p+1}$ of an atom of Ni having $p$ Ni first neighbours, proportional to $[1 - (\Delta U + \rho \delta U)] \chi_{\text{Ni}}$ [5] decreases linearly with $p$: $\theta_{p+1} = \theta_1 - mp$. Using the experimental values for $\theta_1$ and $\theta_2$ we find $\theta_3 = (-1 \pm 10)$ K which means that a Ni atom having two Ni atoms first neighbours is magnetic or very near to be magnetic. Let us assume that it is magnetic and that it induces the maximum moment $\mu_{\text{Ni}}$ on its first neighbours. One can then calculate the saturation magnetization $\sigma_5$ in a model similar to the model which was applied by Boucai et al. [9] to the AuCo alloys. We use the value $\mu_{\text{Ni}} = 2.4 \mu_B$ deduced from our saturation magnetization measurements and from those of Crangle [10]. The calculated values of $\sigma_5$ are much smaller than the measured ones. It seems that the moment per magnetic Ni atom is greater than 2.4 $\mu_B$.

In fact this calculation does not take the effects of the polarization of the matrix by the magnetic impurities into account.

Neutron scattering experiments [11] have demonstrated that as in the case of the PdFe alloys, the magnetic atoms of Ni polarize the matrix at distances as large as 10 Å. But in the case of the PdNi alloys the host is not only the Pd but a PdNi host containing the nearly magnetic Ni atoms and which has a much greater susceptibility than that of the Palladium. Therefore it is possible that the isolated Ni atoms or pairs which lie inside the polarization sphere of a magnetic Ni atom become magnetic. Thus, around each group of three nickel atoms a large induced moment $\mu_{\text{in}}$ may exist, and the moment we have to consider to calculate $\sigma_5$ is not only the moment 3 $\mu_{\text{Ni}}$ of the group but the sum 3 $\mu_{\text{Ni}} + \mu_{\text{in}}$. The existence of that large induced moment is proved by the study of the initial susceptibility $\chi_1$ of the PdNi alloys: For the alloy PdNi 1.5 % for example (Fig. 5), $\chi_1$ measured at $h \approx 18$ Oe between 0.05 and 4 K follows the law:

$$\chi_1 = \chi_0 + C/T$$

$\chi_0$ is due to the non magnetic impurities and the Curie term $C/T$ is due to the magnetic ones. Substracting
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saturation magnetization of the PdNi alloys taking the induced moments into account: We have assumed that an atom isolated, or belonging to a pair has the moment $p_{Ni}$ when it is at a distance $r < R_1$ or $r < R_2$ from a group of three Ni atoms respectively. At distances greater than $R_1$ or $R_2$ the moment is respectively equal to $K_1 \exp(-q r)/r$ and $K_2 \exp(-q r)/r$. The parameters $R_1$, $R_2$, $K_1$, $K_2$ can be deduced from experimental data. The calculation gives good agreement with experiment. It will be published elsewhere.

from the Curie constant $C$ and from $\sigma_0$ the contribution of the parasitic iron atoms ($x = 50$ ppm) associated with a moment of 17.5 $\mu_B$ [13] and from the ratio of these two corrected quantities, we deduce the spin $S$ of the magnetic carriers:

$$S \approx 24$$

which confirms the existence of the giant moments.

The mechanism of the formation of those giant moments has been previously studied in the PdNiFe alloys [13]: one can assume that an atom of iron in the PdNiFe alloys plays a role similar to that of a group of three nickel atoms in the PdNi alloys. The linear increase of the saturation magnetization of the PdNiFe with the iron concentration and the linear increase of the average moment associated with each iron atoms with the Ni concentration (Fig. 6) can be explained by assuming that each Ni atom lying inside the polarization sphere of an iron atom has the saturation moment $p_{Ni} = 2.4 \mu_B$, confirming the fact that the giant moments are due to the polarization of the PdNi host. We have done a calculation of the saturation magnetization of the PdNi alloys taking the induced moments into account: We have assumed that an atom isolated, or belonging to a pair has the moment $p_{Ni}$ when it is at a distance $r < R_1$ or $r < R_2$ from a group of three Ni atoms respectively. At distances greater than $R_1$ or $R_2$ the moment is respectively equal to $K_1 \exp(-q r)/r$ and $K_2 \exp(-q r)/r$. The parameters $R_1$, $R_2$, $K_1$, $K_2$ can be deduced from experimental data. The calculation gives good agreement with experiment. It will be published elsewhere.

Conclusion. — In the PdNi alloys the transition towards the magnetic state is governed by the interactions effects. However they are relatively small since a pair of Ni first neighbours is not magnetic. We have shown that magnetic nickel atoms exist and that they are associated with giant moments. The formation of these giant moments has been investigated in the PdNiFe alloys. These results show that in the PdNi alloys as in a number of other alloys the transition to the magnetic state is not homogeneous.

References