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To cite this version:

HAL Id: jpa-00228730
https://hal.archives-ouvertes.fr/jpa-00228730
Submitted on 1 Jan 1988

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MAGNETIZATION OF VERY DILUTE PdMn AND PdFe ALLOYS: RE-ENTRANT BEHAVIOUR?

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Abstract. — The static magnetization of three very dilute Pd-based alloys containing 700 ppm Fe, 700 ppm Mn and 1 000 ppm Mn has been measured in low applied fields up to 10 Oe as a function of temperature between 50 mK and 1.0 K. The zero field cooled magnetization possesses features which can be replicated by a mean field effective-field model for bond disordered systems with arbitrary spin $S$, suggesting that these alloys may be in the re-entrant regime of the magnetic phase diagram. Measurements of the time dependence of the thermoremanent magnetization yield essentially logarithmic behaviour over a wide range of temperatures.

The observation of apparently re-entrant behaviour in magnetic systems with randomness, characterized by the sequence of transitions from paramagnet-to-ferromagnet-to- a low temperature phase (possibly spin glass), has intrigued both experimentalists and theorists for many years. Such a sequence is predicted by the mean field model of Sherrington and Kirkpatrick [1]; however, the re-entrant phase boundary lies in the regime of replica symmetry-breaking where the solutions of the SK model are unstable [2]. Vector spin models [3] suggest a transition from a collinear ferromagnet to a canted ferromagnetic state accompanied by a freezing of the transverse degrees of freedom.

In both the PdMn and PdFe systems, the percolation threshold for ferromagnetism [4, 5] corresponds to an impurity concentration of approximately 0.1 at %, above which the system response is typically ferromagnetic, and below which the ferromagnetic state gradually disappears. Spin glass behaviour has been observed in the PdFe system [6] below 100 ppm Fe, while PdMn [7] appears to develop spin glass characteristics below 600 ppm Mn, so that both systems possess a potentially re-entrant regime. In the present investigation, we present low field magnetization measurements on three very dilute PdFe and PdMn alloys (700 ppm Fe, 700 ppm Mn, and 1 000 ppm Mn) which have the potential to exhibit re-entrant behaviour, and compare the data with numerical simulations.

The samples were prepared in a conventional inert atmosphere arc furnace by successive dilution of master alloys. The samples were spark out into needles, 0.05 cm in diameter and 1 cm long, and then etched and annealed. The magnetization measurements were performed with a SQUID magnetometer attached externally to the bottom of the copper mixing chamber of a He$^3$/He$^4$ dilution refrigerator. The external magnetic field was applied parallel to the axis of the needles.

Figure 1 shows the temperature dependence of the magnetization of all three alloys measured in a fixed external field $H_a$, all possess a peak and, at higher temperatures, an inflection point below which the ZFC magnetization becomes time dependent. The field cooled (FC) data, obtained after first cooling the sample in the external field $H_a$, coincide with the ZFC data above the inflection point, but increase monotonically with decreasing temperature below the inflection point with $M_{FC}(T) > M_{ZFC}(T)$. At high temperatures, both the FC and ZFC data obey a Curie-Weiss law with an effective giant moment $\mu_{eff} \approx 10 \mu_B$ for both the Mn and Fe impurities in Pd, and with $\theta \approx T_{INFL}$. Figure 2a shows some typical magnetization isotherms for Pd-700 ppm Mn at several temperatures both above and below $T_{INFL} \approx 0.085$ K, in external fields $0 \leq H_a \leq 10$ Oe. An analysis of the initial slope of these isotherms yields an initial susceptibility which "diverges" as $T \rightarrow T_{INFL}$ with an exponent in the external field $H_a$, all possess a peak and, at higher temperatures, an inflection point below which the ZFC magnetization becomes time dependent. The field cooled (FC) data, obtained after first cooling the sample in the external field $H_a$, coincide with the ZFC data above the inflection point, but increase monotonically with decreasing temperature below the inflection point with $M_{FC}(T) > M_{ZFC}(T)$. At high temperatures, both the FC and ZFC data obey a Curie-Weiss law with an effective giant moment $\mu_{eff} \approx 10 \mu_B$ for both the Mn and Fe impurities in Pd, and with $\theta \approx T_{INFL}$. Figure 2a shows some typical magnetization isotherms for Pd-700 ppm Mn at several temperatures both above and below $T_{INFL} \approx 0.085$ K, in external fields $0 \leq H_a \leq 10$ Oe. An analysis of the initial slope of these isotherms yields an initial susceptibility which "diverges" as $T \rightarrow T_{INFL}$ with an exponent

Fig. 1. — Temperature dependence of the magnetization in a fixed applied field. The solid curves represent numerical calculations based on a mean field effective-field model, as discussed in the text.
Table I. - Parameters used to generate “best fit” numerical magnetization curves in figure 1.

<table>
<thead>
<tr>
<th>Alloy</th>
<th>$T_c$ (K)</th>
<th>$S$</th>
<th>$h = \mu_B H_a / k_B T_c$</th>
<th>$\eta = J / J_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pd - 700 ppm Fe</td>
<td>0.470 ± 0.005</td>
<td>5</td>
<td>0.0006</td>
<td>0.93</td>
</tr>
<tr>
<td>Pd - 700 ppm Mn</td>
<td>0.085 ± 0.005</td>
<td>5</td>
<td>0.0017</td>
<td>0.97</td>
</tr>
<tr>
<td>Pd - 1 000 ppm Mn</td>
<td>0.135 ± 0.005</td>
<td>5</td>
<td>0.0010</td>
<td>0.93</td>
</tr>
</tbody>
</table>

SK phase diagram ($0.8 \leq J / J_0 \leq 1.0$). This is illustrated by the solid curves in figure 1, which correspond to numerical calculations of the magnetization for values of the model parameters (listed in Tab. I) which best describe the general features of the experimental ZFC data. Since both the impurity spin $S$ and the reduced field $h$ are predetermined, the only adjustable parameter is $\eta$. The “best fit” values of $\eta$ for all three alloys are re-entrant in the SK sense, and the $\eta$-values for the two PdMn alloys show internal consistency, with the less concentrated alloy located closer to the ferromagnetic-spin glass tricritical point ($J / J_0 = 1.0$), as expected. The remarkable similarity between the experimental and numerical data extends to the magnetic isotherms, as may be seen by comparing the experimental isotherms for Pd-700 ppm Mn in figure 2a with the set of numerical isotherms in figure 2b generated for $\eta = 0.97$, over a similar range of reduced temperatures $\varepsilon = T / T_c$. While the precise location of the re-entrant transition temperature $T_{SG}$ is difficult to determine, model estimates of both the re-entrant SK phase boundary and the AT instability line suggest that $T_{SG}$ is close to the peak in the ZFC magnetization.

Evidence for a re-entrant transition was also sought in the relaxation of the thermoremanent magnetization: the time dependence was logarithmic for observation times $100 \, s \leq \tau \leq 7 \, 200 \, s$, with a weak stretched exponential component at short times. No anomaly was observed in the relaxation rate, which was essentially temperature independent in both systems.


$\gamma \approx 2.0$, somewhat larger than typical ferromagnetic values.

With the inflection point phenomenon tentatively identified with a paramagnetic-ferromagnetic phase transition, with $T_c = T_{INF}$, the presence of the peak in the ZFC magnetization of both systems (and the particularly dramatic decrease in the PdFe magnetization below the peak) suggests the possibility of a further re-entrant transition to a weakly magnetic ground state at lower temperatures. In fact, the behaviour of the ZFC magnetization can be replicated very well by numerical calculations based on a mean field effective-field model [8] for impurities with arbitrary spin $S$ coupled through a Gaussian exchange bond distribution with a mean value $J_0$ and a variance $J$ such that the ratio $\eta = J / J_0$ lies in the re-entrant regime of the