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Neutron Depolarization Studies of Pd-Ni-Fe-P alloy

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Abstract

Bulk metallic glasses based on the quaternary alloy Pd-Ni-Fe-P exhibit interesting phase behavior depending on temperature and applied magnetic field. Previous work has suggested that a range of magnetic phases including paramagnetic, superparamagnetic, ferromagnetic and spin glass can be observed in this system. We have applied one dimensional neutron depolarization to explore the correlation of magnetic moments in $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$ alloy as a function of temperature and applied magnetic field. The results provided evidence for correlation lengths of ~ 200 Å. The nature of the correlations and the formation mechanism of the induced ferromagnetic phase are discussed.

1. Introduction

In the $Pd_{40}(Ni_{1-x}Fe_x)_{40}P_{20}$ metallic glass system, the iron-free alloy is a metallic paramagnet. The addition of Iron to this alloy however generates a range of magnetic phases including paramagnetic, superparamagnetic, spin glass and field-induced ferromagnetic phases as reported for the amorphous alloy of $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$ [1].

The formation mechanism of the field-induced ferromagnetic phase is not yet understood. Hsiao et al. [2] proposed an explanation for this behavior based on competing RKKY interactions between magnetic clusters present in the material. The application of a field overcomes these competing interactions and generates ferromagnetic behavior. These clusters could take the form of nanocrystalline grains within an amorphous matrix or alternatively regions of chemical inhomogenity producing locally coupled magnetic moments. From measurements of the size of the superparamagnetic entities they estimated the size of the nanocrystalline grains to be of the order of 30 - 40 Å. However X-ray diffraction, differential scanning calorimetry, high-resolution TEM and neutron-diffraction measurements indicate that the material is amorphous [3-5], although these techniques may not be sufficiently sensitive to detect smaller magnetic clusters.

We have applied the neutron-depolarization technique to investigate the presence of correlations of magnetic moments in the induced ferromagnetic phase of amorphous $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$.

2. Neutron depolarization technique

The theory of depolarization of polarized neutrons passing through ferromagnetic materials was originally formulated by Halpern and Holstein [6]. Later, the theory was extended to three-dimensional depolarization analyses [7]. The ratio between the neutron polarization P after passing through the sample and the initial polarization P_0 is given by the following equation

$$\frac{P}{P_0} = \left[1 - 2\left\langle \frac{B_\perp^2}{B^2} \right\rangle \left\langle \sin^2\left(\frac{1}{2} cBd\lambda\right) \right\rangle_d \right]^N \qquad (2.1)$$

where B is the total magnetic induction with components of $B_{//}$ and $B_\perp$ with respect to the initial neutron spin direction. d is the average size of magnetic domain/structure in the sample. The number of structures is $N = L/d$ with L being the sample thickness. λ is the neutron wavelength in Å and c = 4.63 x 10^{-6} Å^{-2} T^{-1}.

It is assumed in equation (2.1) that the total induction B of each magnetic structure consists of two components, $B_{//}$ and $B_\perp$. $B_{//}$ is constant throughout the whole sample; while $B_\perp$ is constant within each domain with random directions in the plane perpendicular to $B_{//}$. The factor $\langle B^2_\perp/B^2 \rangle$ in equation (2.1) is a measure of the mean-square magnetic fluctuations in the sample. The correlation length corresponding to the magnetic fluctuations is then represented by d.

3. Experiment

The $Pd_{40}Ni_{22.5}Fe_{17.5}P_{20}$ ribbon for the current experiment was prepared by RF melt spinning with a quench rate of 10^6K/s [4]. The depolarization measurement was performed on the ASTERIX instrument at LANSCE, Los Alamos, USA. The ribbon, with dimensions of about 4 mm wide, 0.02mm thick and 1000 mm long, was wrapped around a 10 mm x 15mm thin aluminum plate. The total thickness of the ribbon in the neutron beam was 3.2 mm. The sample was attached to the cold finger of a closed-cycle refrigerator (CCR) that could produce sample temperatures down to 10K. The CCR was inserted in the core of an electromagnet which provides magnetic fields up to 0.7 T. A polarization cavity polarized the incident neutron beam and a m=3 supermirror polarizer was used to analyze the polarization of the beam after the sample.

The polarization of the transmitted neutrons after the sample was first measured at room temperature and 15 Oe. In this paramagnetic state, the polarization should be unaffected by the sample and hence was used as a measure of the initial-beam polarization [8]. The sample was then cooled to 10 K under the same 15 Oe field. At 10 K the polarization was the same as at room temperature within experimental error. This is as expected, as the sample should be in the spin-glass state. In this state the variation in the magnetic fluctuations are too quick for the neutron to precess and so no change in the neutron beam polarization should be observed [8]. Subsequently, the sample temperature was set at 30 K, corresponding to the centre of the induced ferromagnetic state as reported by Shen *et al.* [1]. The polarization was then measured as a function of neutron wavelength at several applied magnetic fields.

4. Results and discussion

The measured depolarization data, as a function of neutron wavelength, are presented in figure 1. It shows a small but discernable depolarization that increases with increasing wavelength. The lines in the figure are fits to the experimental data.

In principle, fitting equation (2.1) to the experimental depolarization data provides values for the mean-square magnetic fluctuation and the correlation length. However, a one-dimensional depolarization experiment cannot give unique and independent solutions for these two quantities. Taking magnetization data measured at 30 K and

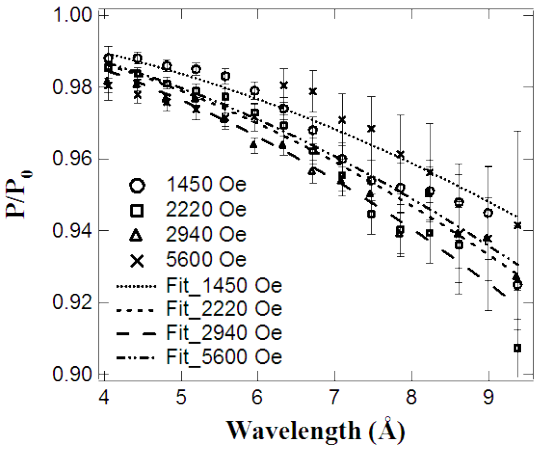

Figure 1. Depolarization as a function of wavelength for several applied magnetic fields

assuming that, at fields above 1 kOe all magnetization changes were associated with rotation of the magnetization away from the easy axis, we calculated the magnetic induction and its perpendicular component for each field in Figure 1. The resulting correlation lengths determined from fitting the data ranged from 160 Å to 264 Å showing an increase trend with increasing magnetic field below 3 kOe and no significant increase for fields above 3 kOe, as shown in figure 2.

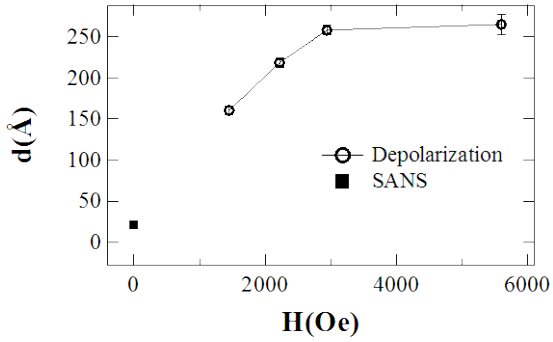

Figure 2. Correlation length as a function of applied magnetic fields

It is interesting to note that a previous small-angle neutron scattering measurement on the same sample at 33 K and zero field obtained a correlation length of 21 Å as indicated by the square symbol in figure 2 [5]. The SANS datum is reasonably consistent with the depolarization data in terms of field dependence.

Combining the neutron depolarization data with the SANS datum, we may postulate a model of interacting clusters of magnetic moments for the field induced ferromagnetic phases of the PdNiFeP system. The zero field correlation length measured by SANS may be associated with the chemical inhomogenities suggested by Hsiao et al [2]. These chemical inhomogenities contain magnetic clusters. Upon applying magnetic field, larger magnetic structures are formed from these magnetic clusters in a Pd-rich matrix. The larger correlation lengths, observed by neutron depolarization, may correspond to the correlations of groups of large magnetic structures having relatively uniform magnetization vectors. This clustering of magnetic entities to form larger structures has been previously observed in nanocrystalline ferromagnets in the form of interaction domains [9,10].

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