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Magnetic and anomalous magnetic viscosity in the bulk amorphous ferromagnet $Pr_{58}Fe_{24}Al_{18}$ and partially amorphous ferromagnet $Pr_{60}Fe_{24}Al_{16}$

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ABSTRACT

A number of ferromagnetic Pr–Fe–Al alloys have been prepared by argon arc melting and quenching into a copper mould. The alloy of composition $Pr_{58}Fe_{24}Al_{18}$ is identified as being amorphous (bulk metallic glass or bulk amorphous ferromagnet), and a range of magnetic measurements have been performed to explore differences and similarities between it and a partially amorphous alloy, containing a significant crystalline fraction, $Pr_{60}Fe_{24}Al_{16}$. For both alloys, measurements of the irreversible susceptibility, and magnetic viscosity on the major hysteresis loop are reported. From the magnetic viscosity data, the fluctuation field is determined. The behaviour of the anomalous magnetic viscosity (non-monotonic behaviour of the magnetic viscosity, where the magnetisation as a function of time is seen to increase, reach a peak, and then decrease), on the recoil curve that leads to the dc demagnetised state is investigated. Both alloys display non-monotonic behaviour. After dc demagnetisation, spontaneous remagnetisation is observed in both alloys, and some comments are made on the thermal remagnetisation behaviour of the amorphous alloy. The anomalous magnetic viscosity is interpreted in the context of the Preisach model, as it predicts a simple functional relationship between the time taken to reach a peak and the applied magnetic field. The experimental data for both alloys is in good agreement with this prediction.

1. Introduction

Time-dependent behaviour of the magnetisation is a well known feature of ferromagnetic materials, and arises as thermal activation processes transform metastable states to stable states [1]. The exact nature of the time-dependent magnetisation is a consequence of the magnetic history that the material may have been subject to. The magnetic history for observing magnetic viscosity, where the magnetisation is seen to vary in a monotonic fashion as a function of time, is a *One-Step* process. Here the magnetic field is applied in the forward direction to achieve magnetic saturation of the material, then ramped in a negative direction to a point on the major hysteresis loop in the second or third quadrant and held at a constant value. Over limited time scales, the time dependence of the magnetic polarisation $J(t)$ follows the well known expression [1]

$$J(t) = S\ln(t) + \text{const}, \tag{1}$$

where S is the magnetic viscosity parameter. A related parameter, often determined from magnetic viscosity data, is the fluctuation field, H_f, which is defined by S/χ_{irr}, where χ_{irr} is the irreversible magnetic susceptibility [2,3]. H_f can be viewed as a measure of thermally activated rate processes that govern magnetisation reversals.

Anomalous magnetic viscosity manifests itself as the non-monotonic behaviour of the time-dependent magnetisation, where the magnetisation is seen to increase, reach a peak, and then decrease, following a *Two-Step* (or double field reversal) magnetic field history. In this case, rather than holding the applied field constant at a point on the major hysteresis loop, as is done in the *One-Step* process for observing magnetic viscosity, a second step is incorporated, with the point on the major hysteresis loop becoming a turning point. From the turning point, the applied field is ramped in a positive direction along a recoil curve, to a point where the applied field is held at a constant value and the magnetisation measured as a function of time. Of special interest is the behaviour of the anomalous magnetic viscosity along the recoil curve that leads to the dc demagnetised state. The dc demagnetised state is reached by choosing a turning point on the major hysteresis loop to give a recoil curve that terminates at the origin of the M–H plane, i.e. when the applied field is zero the magnetisation is zero. From the dc demagnetised state, if the applied field is maintained at zero and the temperature constant, the magnetisation will be seen to increase with time. This is the phenomenon of spontaneous remagnetisation. Related to this effect is thermal remagnetisation, where again the applied field is

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maintained at zero, and as the temperature is increased the magnetisation is seen to increase.

Anomalous magnetic viscosity was initially observed in a melt-spun $Nd_4Fe_{77}B_{19}$ magnet [4] and metal particle thin film recording material [5], but the effect is small, and its study was inhibited, to some extent, by the limited sample mass possible from glassy metal ribbons or thin films, even when coupled with the most sensitive SQUID magnetometer. More recently anomalous magnetic viscosity has been observed in the bulk amorphous ferromagnets $Nd_{60}Fe_{30}Al_{10}$ [6,7] and $Nd_{60}Fe_{20}Co_{10}Al_{10}$ [8]. The advent of bulk metallic glasses (BMG) or bulk amorphous alloys [9], particulary (Nd,Pr)–Fe–Al alloys which possess hard magnetic properties (termed bulk amorphous ferromagnets), has resulted in a material that seems to be ideal for studying time-dependent magnetic effects [10–13]. Unlike glassy metal ribbons which are only a few microns in thickness, and require cooling rates of 10^6 K/s, bulk amorphous alloys form at much more modest cooling rates, typically a few tens K/s, and can be cast into rods some millimetres or even centimetres in diameter [14].

Time-dependent behaviour of the magnetisation in Pr–Fe–Al alloys has not been studied previously. In the study reported here, a range of Pr–Fe–Al alloys have been prepared, and two alloys, $Pr_{58}Fe_{24}Al_{18}$, which is a bulk amorphous ferromagnet, and $Pr_{60}Fe_{24}Al_{16}$, which is partially amorphous, were selected for study. A fully amorphous alloy and a partially amorphous alloy, the latter containing a significant crystalline fraction, were chosen to explore the impact, if any, of crystallinity on the time-dependent magnetic behaviour. For both alloys, the magnetic viscosity was investigated following a *One-Step* process, and the anomalous magnetic viscosity following a *Two-Step* process. From the magnetic viscosity data the fluctuation field, for each alloy, was determined. The anomalous magnetic viscosity was investigated for a range of applied fields along the recoil curve that leads to the dc demagnetised state. The behaviour of the irreversible susceptibility is reported for both alloys. Measurements of the spontaneous remagnetisation in both alloys are presented, along with measurements of the thermal remagnetisation in the amorphous alloy, $Pr_{58}Fe_{24}Al_{18}$. The Preisach model is found to provide a useful framework for understanding the anomalous magnetic viscosity, spontaneous remagnetisation and thermal remagnetisation. In the case of anomalous magnetic viscosity the Preisach model predicts a simple functional relationship between the time taken to reach a peak and the applied magnetic field, and the experimental data, for both alloys, is in good agreement with this prediction.

2. Experimental procedures

2.1. Synthesis and structural characterisation

A range of Pr–Fe–Al alloy samples were prepared by argon-arc melting and suction casting into a split copper mould. Each sample was typically in the form of a rod about 6 mm long and 2 mm in diameter. X-ray powder diffraction (XRD) with Cu$-K_\alpha$ radiation was used to structurally characterise each of the alloy samples, and it was on the basis of the XRD patterns that a fully amorphous and partially amorphous (or partially crystalline) specimen were selected for the time-dependent magnetic measurements.

It was found that the alloy of composition $Pr_{58}Fe_{24}Al_{18}$ was amorphous. Its XRD pattern is shown in Fig. 1(a) and it can be seen that there are diffuse broad Bragg peaks centered around 2θ values of 30° and 55°, which is consistent with a structural coherence length of <5 nm [13]. Saito [12] has undertaken a study of the Pr–Fe–Al phase diagram, and reports a similar XRD pattern

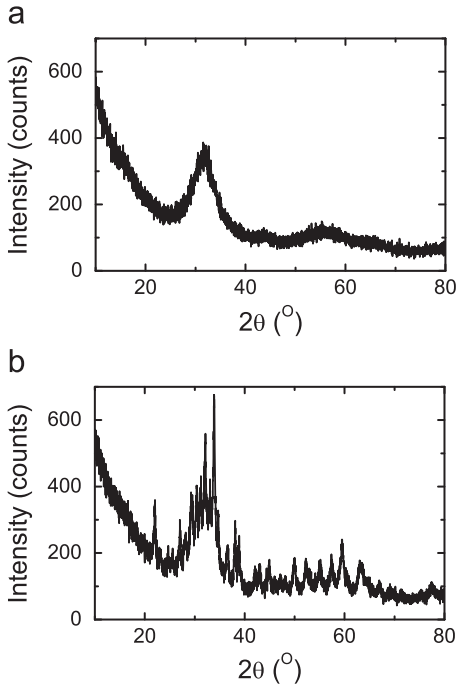

Fig. 1. X-ray diffraction patterns of (a) the amorphous alloy $Pr_{58}Fe_{24}Al_{18}$ and (b) the partially amorphous alloy $Pr_{60}Fe_{24}Al_{16}$.

for a Pr–Fe–Al bulk amorphous alloy, as does [13] for bulk amorphous Nd–Fe–Al. It has been pointed out by McCallum et al. [13] that laboratory XRD lacks resolution for revealing fine structural detail, that may have a nanometre length scale. It has been generally accepted that if a bulk amorphous alloy has a XRD pattern similar to that shown in Fig. 1(a), then it is amorphous. However, when examined by techniques that have a higher resolution, for example those using synchrotron radiation, structure on the nanometre scale is revealed. High resolution synchrotron based techniques have been used to probe Nd–Fe–Al bulk amorphous alloys and reveal the existence of small crystalline clusters of approximately 1.2 nm, embedded within the amorphous matrix [13]. (To the best of our knowledge, there have been no comparable microstructural studies made on Pr–Fe–Al bulk amorphous ferromagnets.) What is termed bulk amorphous, is perhaps better viewed as a collection of nanocrystalline phases or entities embedded in an amorphous matrix, but which appear to be amorphous based upon laboratory XRD.

It was found that an alloy of composition $Pr_{60}Fe_{24}Al_{16}$ was partially amorphous, containing a significant crystalline component. Its XRD pattern is shown in Fig. 1(b), and it shows quite clearly a series of well defined sharp Bragg peaks, indicating the presence of crystalline phases. By correcting the XRD spectra, of both the amorphous and partially amorphous alloy, for the diffuse background scattering and analysing the structure of the various peaks, we estimate the alloy to be approximately 60% amorphous and approximately 40% crystalline.

2.2. Magnetic characterisation

All magnetic measurements, with the exception of the thermal remagnetisation measurements, were performed at room temperature (294 K) using a lake shore vibrating sample magnetometer mounted on an electromagnet, capable of applying magnetic fields of up to 2 T. The thermal remagnetisation measurements were made in the range of 293–400 K using a