

Determination of Disordered Magnetic Structures in High-Coercivity Nd-Fe-Based Glassy Alloys

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Binary rare earth - Fe amorphous alloys have been extensively studied in the past not only for their physical interest but also for technological importance for permanent magnets, magnetostrictive materials and magneto-optical recording media. These alloys form a variety of magnetic structures ranging from ferromagnetic to sperromagnetic and sperrimagnetic, depending on the relative magnitude of the random magnetic anisotropy (RMA) with respect to the exchange energy [1,2]. The high coercivities observed by Croat [3] in melt-spun rare earth - Fe (rare earth = Nd or Pr) alloys was the first evidence for the existence of an unknown metastable hard magnetic phase in the light rare-earth - transition metal binary amorphous system. More recently, it has been reported that Nd-Fe-(Si,Al) ternary amorphous alloys are formed in a wide range of compositions in different bulk shapes and exhibit large coercive fields at room temperature [4,5].

In this work, we focus on the determination of magnetic structures in high-coercivity $\text{Nd}_{90-x}\text{Fe}_x\text{Al}_{10}$ ($x = 20 - 60$ at. %) bulk amorphous alloys using RMC modeling of neutron diffraction data, Mössbauer spectra analysis and specific magnetic measurements. We measured both melt-spun ribbons with thicknesses between 25 and 120 μm and cast rods with diameters no larger than 2 mm.

Neutron diffraction measurements have been made at 12 temperatures between 15 and 773 K. The data have been modeled using the reverse Monte Carlo method (RMC) [6] under the assumption that the atomic structure is completely disordered (Figure 1). From the resulting configurations we find that the near-neighboring ordering is asperomagnetic, but the magnetic structure is predominantly ferromagnetic at temperatures below crystallization temperature (about 440°C) in agreement with the static magnetic measurements [7] and Mössbauer spectra (Figure 2). The pair correlation function obtained by Fourier inversion of the diffraction patterns shows first neighbor peaks at 2.54, 2.85 and 3.36 Å. The peak at 2.54 Å is at the position expected for the nearest neighbor Fe atoms in the dense random packing (DRP) model; the other peaks cannot be correlated to combinations of the radii of Nd (1.82 Å), Al (1.43 Å) or Fe (1.27 Å) atoms as predicted by the DRP model. This disagreement could be ascribed to the development of a new type of disordered structure. Thus, one suggests that the structure consists in a dense random packing of nanometer-sized atomic clusters. Consequently, the magnetic structure will consist in non-collinear short-range magnetic ordered regions randomly distributed.

The granular magnetic structure consists in Fe-based nanosized clusters dispersed in amorphous Nd-rich matrix. Due to their very small dimensions they cannot be evidenced by X-ray diffraction measurements, but their presence is proved by the specific magnetic behavior, especially by the high coercive fields (up to 2 T) in the as-quenched state and the dependence on the cooling rate. The composition and size of the magnetic clusters as well as the composition of the matrix in which the clusters are embedded are very sensitive to the preparation conditions, mainly the cooling rate and the thermal history of the molten alloy.

Fe-based magnetic clusters are coupled between them through the amorphous matrix and the exchange coupling is strongly influenced by the magnitude of the Nd^{3+} single ion local anisotropy. Whereas at low temperatures the anisotropy plays the predominant role in the macroscopic magnetic response of the system, the increase of the temperature diminishes its effect and the main role is attributed to the ferromagnetic exchange energy. At low temperature, the local anisotropies of the Nd^{3+} ions are very strong and oriented randomly, and the local magnetic moments of Fe and Nd are “frozen” randomly. The anisotropy axis corresponding to the Fe-Nd pairs are oriented randomly, and a small external field is not enough to align them. The maximum of the coercive field is attained at that temperature at which the anisotropy energy and the exchange energy have similar values, i.e. the magnetic clusters have the size of the single magnetic domain (smaller than 100 nm). As larger the applied field as smaller the temperature at which the coercive field attains its maximum.

Despite of that, there are still many questions related to the microstructure of these materials and its interplay with magnetic properties. The magnetic ground states of Nd-Fe-based cluster amorphous alloys and non-collinear structures existent in these materials are far from being fully characterized. These problems and others make the study of the magnetic properties of bulk amorphous alloys a fascinating field of research.

References

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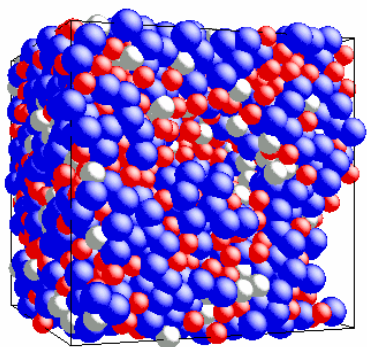


FIG. 1. Modeled atomic structure for amorphous $\text{Nd}_{50}\text{Fe}_{40}\text{Al}_{10}$ melt-spun ribbon, 25 μm in thickness

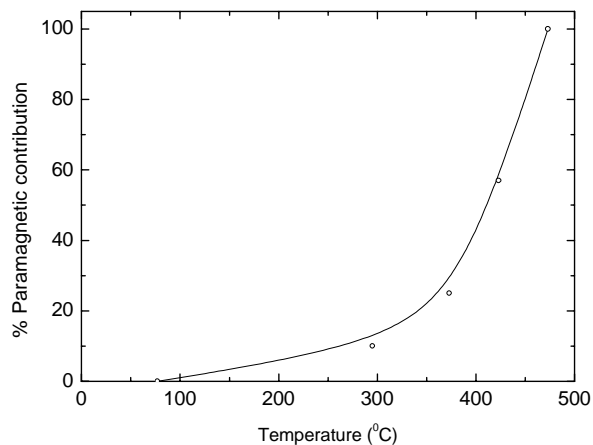


FIG. 2. The evolution of the paramagnetic fraction vs. temperature for $\text{Nd}_{50}\text{Fe}_{40}\text{Al}_{10}$ bulk amorphous alloys