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ATOMIC ORDERING AND MAGNETIC PROPERTIES IN $Nd_{50}Fe_{40}Al_{10}$ GLASSY HARD MAGNETS

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Structures with atomic medium-range order (MRO) of 2-4 nm are observed in the $Nd_{50}Fe_{40}Al_{10}$ "amorphous" melt-spun ribbons by high resolution transmission electron microscopy (HRTEM). The volume fraction and size of the MRO regions increases as the cooling rate decreases, leading to the increase of the coercive field from 80 kA/m to about 350 kA/m when the ribbon thickness increases from 25 to 120 μm. The huge increase of the coercive field up to 4 MA/m as temperature decreases below room temperature in the maximum applied field of 7.2 MA/m is ascribed to the competition between exchange interactions and local anisotropies. The role of the MRO regions as pinning centers is discussed.

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1. Introduction

The magnetism of light rare earth – Fe based alloys produced by rapid quenching from the melt is a great stimulus for research in their preparation and behavior, especially since the discovery of the high coercive fields over 10 kOe at room temperature in melt-spun Nd-Fe and Pr-Fe amorphous alloys [1-3]. Their magnetic behavior is strongly related to the magnetic atoms environments and the attempts trying to explain this specific behavior were not always consistent with each other. The most used hypothesis was that of the presence of two or more magnetic phases. Recently, the similar magnetic behavior was reported for (Nd,Pr)-(Fe,Co)-(Al,Si) amorphous alloys [4-12]. Moreover, these alloys can be cast in bulk shapes with dimensions up to a few millimeters due to their large glass-forming ability.

In this paper, the effect of the structural disorder on the magnetic properties of $Nd_{50}Fe_{40}Al_{10}$ melt-spun ribbons with thicknesses varying between 20 and 120 μm is investigated systematically in the temperature range 4 – 800 K. Possible mechanisms for the huge coercive field observed due to the presence of medium-range ordered regions are discussed.

2. Experimental

$Nd_{50}F_{40}Al_{10}$ melt-spun ribbons with different thicknesses were prepared by single roller melt-spinning method in an Ar atmosphere by changing the wheel velocity from 30 m/sec to 2.5 m/sec. X-rays, neutrons and high energy synchrotron X-rays diffraction patterns confirmed that all investigated ribbons exhibit disordered structures. The only features not necessarily characteristic to the conventional amorphous alloys are the split of the first diffraction peak and the pronounced structure at high angles indicating the presence of short or medium –range ordered regions within

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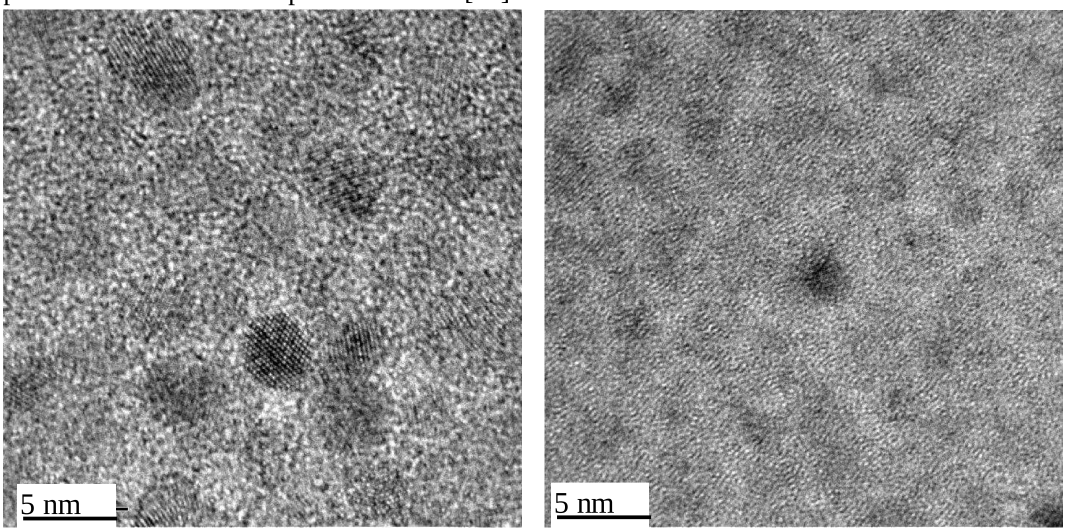
the amorphous matrix [12]. Atomic scale investigations were performed by means of high resolution transmission electron microscopy (HRTEM) after the specific preparation of the investigated surfaces. The images were taken using a 300kV electron microscope. Magnetic measurements in the temperature range 4-800 K were carried out using an Oxford MagLab VSM in maximum applied fields of 7.2 MA/m. The magnetic field was applied in the axial direction of the samples. To obtain information about either the distribution of moments due to chemical non-equivalency of the magnetic atoms or about the distribution of moments between Fe and Nd magnetic atoms, Mössbauer measurements were performed between 77 and 473 K in transmission geometry.

3. Results and discussion

3.1. Atomic ordering

Figure 1(a) and 1(b) show HRTEM images taken from very small area of $Nd_{50}Fe_{40}Al_{10}$ melt-spun ribbons of 25 and 120 μm, respectively. Medium-range ordered (MRO) regions of about 2-3 nm having mainly fcc structures, the nominal composition $Fe_{77.2}Nd_{22.8}$, and the lattice fringe spacing of about 0.25 nm frequently appear in thin ribbons of 25 μm and, in addition the fringe geometries are randomly distributed within the amorphous matrix and cross each other sometimes. Our previous neutron diffraction studies indicated that the Fe atoms have the Fe and Nd nearest neighbors at 0.254 nm and 0.285 nm, respectively [12]. The structure is different for the thick ribbons of 120 μm showing over the whole area a modulated contrast consisting of fringes with a size of about 0.5 nm typical for the relaxed amorphous structure [13].



(a) (b)

Fig. 1. Bright-field electron micrograph of the as-cast $Nd_{50}Fe_{40}Al_{10}$ melt-spun ribbons: (a) 25 μm, and (b) 120 μm thickness.

The difference between the 2 images is predictable, as being caused by the decrease of the cooling rate during the melt-spun amorphous ribbons preparation process. The average size and the volume fraction of the fcc MRO clusters increases as the quenching rate decreases, i.e. the ribbons thickness increases, leading to a higher percolation limit.

3.2. Magnetic Properties

The structural results clearly show the presence of two phases in $Nd_{50}Fe_{40}Al_{10}$ melt-spun ribbons, regarding of their thickness: the fcc clusters rich in Fe, which are embedded in the amorphous matrix rich in Nd. In agreement with our previously reported results [14], anew type of disordered structure is revealed in $Nd_{50}Fe_{40}Al_{10}$ amorphous alloys, which consists of dense random packing of nanometer-sized atomic clusters, whose size is dependent on the samples thickness, i.e. the quenching rate. Consequently, one expects complex magnetic behavior dependent on the atomic arrangements, temperature and preparation conditions.

The variation of the magnetization and coercive field on temperature for $Nd_{50}Fe_{40}Al_{10}$ melt-spun ribbons of 25 and 120 μm, for different applied fields is presented in Fig. 2(a) and Fig. 2(b). For both types of ribbons the maximum of the coercive field and magnetization moves towards lower temperatures when the applied field increases.

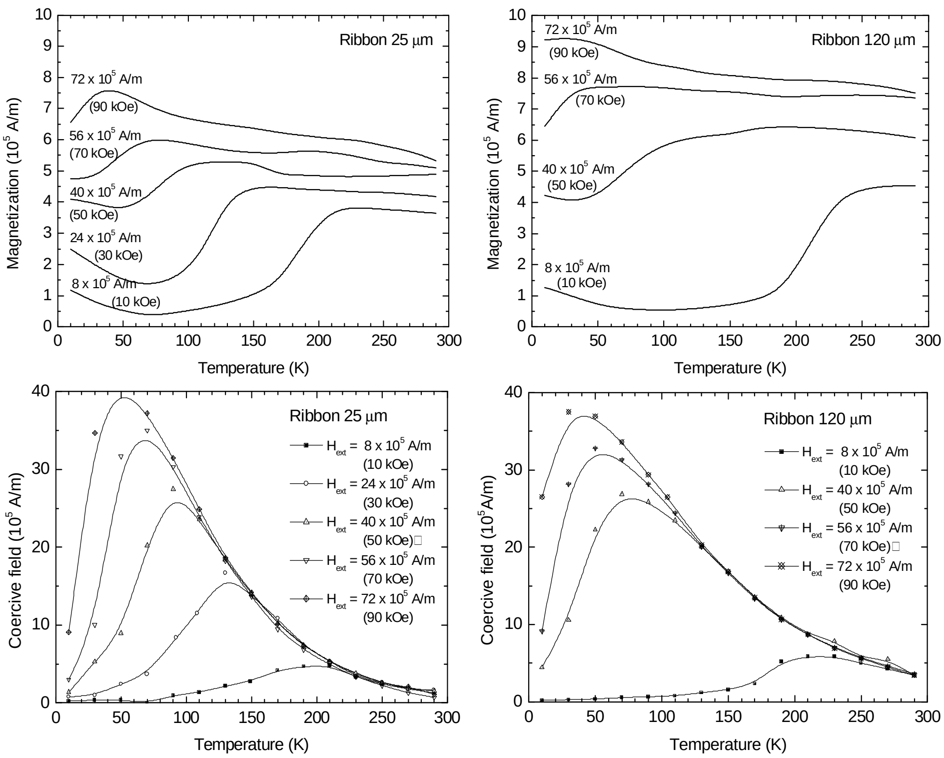


Fig. 2. Magnetization and coercive field vs. temperature for amorphous melt-spun ribbons $Nd_{50}Fe_{40}Al_{10}$.

At high temperatures (around room temperature) the magnetic Fe atoms participate in the reversible magnetization along with some paramagnetic contribution. Mössbauer data are consistent with this (Fig. 3).

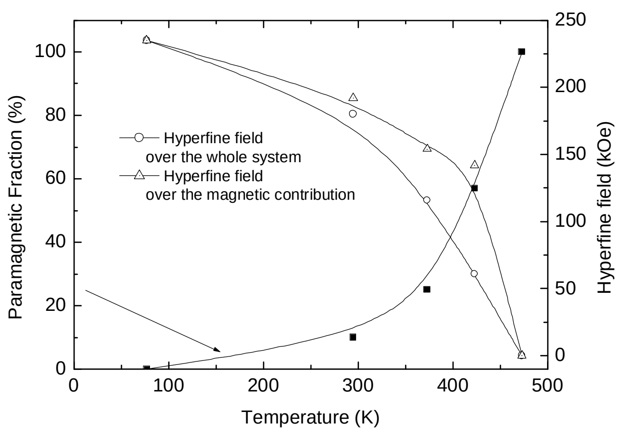


Fig. 3. Hyperfine fields and the evolution of the paramagnetic fraction vs. temperature as resulted from Mössbauer data.

The Nd atoms in the Fe-rich region (fcc patches in Fig. 1), may order speromagnetically and have no magnetic contribution. As the temperature is lowered the thermal effects are weaker and the coercive field increases (Fig. 2(b)). At a critical temperature, ranging from 30 to 210 K depending on the external field value, some Fe atoms within the MRO regions become pinned so strongly in orientation that the available applied field cannot reorient them along the field direction. Thus as the temperature is lowered, more and more Fe moments become frozen and they have no contribution to the magnetization processes. Since the material has a paramagnetic contribution to the magnetization, the value of the magnetization is small and positive when temperature is below the critical temperature (Fig. 2(a)).

Above room temperature the paramagnetic fraction corresponding to the Nd-rich matrix increases and the magnetic moment of the Fe atoms decrease due to the frustration of the ferromagnetic exchange interactions by the thermal effects, as shown in Fig. 3. The magnetic disorder is attained at 473 K.

4. Concluding remarks

Despite the considerable progress achieved in recent years concerning the knowledge of the atomic structure of amorphous alloys and concerning the understanding of their basic magnetic properties, several unsolved problems were encountered in the field of light rare earth (Nd, Pr)-TM amorphous alloys. 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-Al clustered amorphous alloys and non-collinear structures existent in these materials are far from being fully characterized. These features incite for deeper investigations being a very fascinating field for knowledge research.

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