Low Temperature Magnetic Properties of Nanocrystalline Iron

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Abstract. Nanocrystalline iron has been obtained by sputtering on different substrates. Resistivity measurements point out an anomalous behaviour of the thermal dependence of the resistivity at low temperatures which dissapears when a high magnetic field is applied. Strong changes in the spin dependent scattering in islanded Fe (110) thin films are due to a low temperature spin freezing of the island boundary magnetic regions, producing a suppression of the exchange coupling between islands. A consequence of the magnetic decoupling is the random arrangement of the individual magnetization, determined by the magnetocrystalline anisotropy of each island, which results in a spin-dependent induced increase of resistivity below freezing temperature. After application of a magnetic field, magnetic ordering recovers regardless of temperature, obtaining a typical metallic behavior for the temperature dependence of the resistivity. Ratio of low temperature resistivities, measured with and without magnetic field, yields magnetoresistance values of up to 5.5% for 16.5% nm in-plane island sizes. The following two conclusions are obtained, i) nanocrystalline Fe can be considered a low temperature GMR-like system and ii) Fe grain boundaries are not ferromagnetic at low temperature, but behave as reentrant spin-glass.

9.1 Introduction

There exists a great interest in the development of nanostructured magnetic materials due to novel behaviors observed. When the size of the magnetic entities is reduced to the nanoscale range, the structural correlation length becomes of the same order as the exchange correlation length, thus giving rise to new phenomena. In particular, the relative number of atoms located at the grain boundaries increases drastically. Since magnetic properties are closely linked to short range order that fluctuates at the boundaries, differences in the magnetic behavior of grain and grain boundaries could be expected. In this work, the evolution of the magnetic properties of nanocrystalline Fe grown by sputtering with different in-plane and out of plane dimension is shown. We present the low temperature magnetic behavior of this system by measurements of transverse Kerr effect, the thermal dependence of the magnetization and electrical resistance. These measurements evidence the existence of a low temperature magnetic disordered state at the grain boundaries.
Nanostructured magnetic materials have attracted a lot of interest since their macroscopic magnetic properties are very different from those of the bulk. These materials are formed by an assembly of magnetic grains, called nanocrystals, embedded in a magnetic or non-magnetic matrix. Nanocrystals are generally interconnected by either the grain boundaries or a different matrix. As the size of the crystallites is reduced, the number of atoms located at the grain boundary becomes comparable to the number of atoms on regular lattice sites. A simple calculation reveals that when the mean grain size \( D \) is around 10 nm, the fraction of atoms at the interface between the crystallites can be as high as 30%. Finite size and interface effects are claimed to be the main responsible for the novel physical properties exhibited by bulk nanocrystalline materials.

Let us focus on ferromagnetic nanocrystals. Generally we mean by ferromagnetic nanocrystalline sample any sample composed of ferromagnetic nanocrystals embedded in a matrix. The relevant aspect of magnetic nanocrystals is related to the coincidence of the grain size scale (nanometers) with the critical magnetic length. One important characteristic of nanocrystalline ferromagnetic samples is the orientation fluctuation of the local magnetization easy axes. The correlation length of the anisotropy axes is typically of the order of \( D \). The second important characteristic is the degree of coupling, exchange or magnetostatic, between adjacent crystallites carried out through the atoms of the intergranular region which in general exhibit different magnetic properties from the atoms place inside the nanocrystals. The difference between exchange and magnetostatic coupling is remarkable. Exchange is more intense and localized, whereas dipole-dipole interactions are smaller but long ranged. Exchange from ferromagnetic toward paramagnetic metals can penetrate a few interatomic distances decaying exponentially. But its boundary value at the ferromagnetic side reaches effective fields of 1000 T. Exchange interactions also govern the coupling through insulators driving the spin dependence of electronic tunnelling. On the other hand, the stray fields carrying magnetostatic coupling are of the order of a few teslas but extend over the overall crystallite size. Both types of interaction lead to ordered ground states at low temperatures. The ground state is in general difficult to predict since many types of interaction are present.

From the point of view of their practical uses, coercivity is by far the most relevant parameter characterizing the behaviour of nanostructured magnetic materials. The coercivity is firstly related to the intrinsic properties of the main phase forming the material. The central intrinsic property influencing coercivity is the magnetic anisotropy giving the magnitude of the maximum energy barrier to be overcome during the magnetization reversal. Other intrinsic quantities influencing the field at which the global magnetization reverses are the saturation magnetization, \( M_s \) and the order temperature.

The influence of the nanostructure on the macroscopic anisotropy of a ferromagnetic solid is a beautiful example of the dependence of properties on the ratio between structural lengths and characteristic magnetic lengths. In a ferromagnetic homogeneous specimen the following characteristic lengths are normally defined: