DIRECT EVIDENCE OF THE EXISTENCE OF CORRELATED NANOMETRIC MAGNETIC GRAINS IN Fe-BASED AMORPHOUS AND CRYSTALLINE ALLOYS BY SANS

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There is growing experimental evidence that (i) magnetic inhomogeneity or the so-called ‘magnetic microstructure’ is an attribute that is *inherent* to magnetic systems as different as amorphous (or crystalline) ferromagnets, nanocrystalline soft magnetic alloys, nanostructures, fine ferromagnetic particles, granular giant magnetoresistance (GMR) materials, colossal magnetoresistance (CMR) manganates and frustrated pyrochlore oxides, and (ii) the nature of magnetic inhomogeneity basically decides the magnetic behaviour of a given system [1]. In this sense, we have synthesized two types of alloys, the FeCuAg nanocrystalline granular alloys, $Fe_{13}Cu_{10}Ag_{77}$ and $Fe_{22}Cu_{14}Ag_{64}$ [2], and an amorphous alloy of $Fe_{91}Zr_9$ [3]. In the FeCuAg case, the system is constituted by Fe(Cu) magnetic nanometric grains in a diamagnetic Ag matrix. These alloys present an *interacting* superparamagnetic behaviour with the existence of dipolar interactions among the Fe(Cu) grains. The intensity of the interactions increases when the concentration of magnetic grain diluted in the matrix increases, as it is observed when going from $Fe_{13}Cu_{10}Ag_{77}$ to $Fe_{22}Cu_{14}Ag_{64}$. However, the $Fe_{91}Zr_9$ alloy is formed by ferromagnetic clusters in a ferromagnetic matrix and behaves as a re-entrant ferromagnet. For the characterisation and analysis of the inhomogeneities in these systems and of how they are responsible of the observed magnetic behaviour, the alloys have been studied by Small Angle Neutron Scattering (SANS). This is a unique technique to characterise *directly* magnetic inhomogeneities of sizes from 1 to 1000 nm. Measurements have been carried out at different temperatures and it has been possible to separate the magnetic and nuclear contributions to the total scattering intensity by applying a large enough magnetic field to saturate the samples. From the magnetic component to the intensity is possible to obtain values for the size distribution of magnetic grains/clusters embedded in the matrix.

In the case of the Fe-Cu-Ag alloys, the SANS data show no evidence of changes in the size of the nanometric magnetic grains with temperature above and below the blocking temperature. The signal corresponds to particles in different environments, most of them (95 vol. %) are small single-domain (approx. 4 nm) superparamagnetic particles (low degree of interparticle interactions), and there is a minor fraction, in which the grains are larger (< 14 nm) and will present enhanced magnetic correlations among the grains. Respect to the $Fe_{91}Zr_9$ alloy, the SANS intensity displays important changes for different temperatures (10, 90, 160, 300 K), characteristic of a re-entrant spin-glass state for low temperatures ($T < 50$ K), ferromagnetic behaviour ($50 \text{ K} < T < T_C = 210$ K), and paramagnetic for the pattern at RT, respectively. In this alloy two types of clusters coexist, on one hand, 2 nm clusters *completely isolated* from the ferromagnetic matrix and, on the other, *partially isolated* clusters that increase their size from 2 nm when increasing temperature through two processes: merging together because of the strong coupling between the neighbouring clusters to form a bigger cluster or polarizing an increased number of spins originally belonging to the FM matrix via direct exchange interactions. However, the temperatures in excess of T_C disorder not only the ferromagnetic matrix but also the clusters and hence the cluster size decreases for temperatures above T_C.

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