Role of interface in the interparticle magnetic interactions in Fe_xAg_{100-x} ($20 \leq x \leq 55$) granular thin films

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Nanogranular solids present a rich variety of magnetic states, which are related to, among other factors, the magnetic interactions. These interactions depend on the size and the separation of the magnetic nanoparticles, but also on the nature of the nanoparticles/matrix interface, as seen in Fe-Ag and Co-Cu granular thin films [1-3]. In order to study the role of this interface, we have prepared Fe-Ag thin films (~100-200 nm) in the range of 20-50 at. % Fe by pulsed laser deposition technique (PLD). These films have been deposited at 300 K onto Si(100) substrates, and coated with ~10 nm of Ag. Their compositions have been determined by energy dispersive X-ray analysis (EDX) and the structure has been studied by X-Ray Diffraction (XRD), High Resolution Transmission Electron Microscopy (HRTEM), Grazing Incidence Small Angle X-Ray Scattering (GISAXS) and X-Ray Absorption Spectroscopy (XAS). DC and AC zero-field-cooled/field-cooled (ZFC-FC) curves have been measured, using SQUID and PPMS magnetometers, as a function of temperature (5-350 K) and the applied magnetic field (5-100 Oe).

Structure analysis has revealed that Fe_xAg_{100-x} are basically composed of small bcc Fe nanoparticles (~2-3 nm) inside a granular Ag fcc matrix (~10 nm). For the $Fe_{50}Ag_{50}$ sample, together with these crystalline nanoparticles, an interparticle amorphous $Fe_{50}Ag_{50}$ alloy has been found, with a variable thickness of a few nm, as revealed by ultra-HRTEM micrographs (see figure 1, left). This interface occupies around a 25 % of the sample, as indicated by EXAFS fittings. As the Fe concentration decreases, the system becomes more amorphous like.

Concerning the magnetic behaviour, $Fe_{50}Ag_{50}$ sample shows two magnetic transitions at high and low temperatures (see figure 1, right). We have determined that these transitions are mediated by the magnetic ordering/disordering of the amorphous interface.

At low temperatures (T < 65 K), this interface is in a spin glass like state, as corroborated by the presence of an exchange bias phenomenon in the hysteresis cycles *M(H)*. Consequently, the amorphous interface disables the transmission of the direct exchange interactions between the Fe nanoparticles. This makes the global magnetization decrease, and the system enters into a superspin glass (SSG) state, mediated by the dipolar interactions, the intraparticle anisotropies, and the inherent structural disorder. If the magnetic field increases, the spin glass disorder is rapidly overcome. Using a modified random anisotropy model [4], we have estimated that the effective anisotropy of the Fe nanoparticles is $K = 0.9.10^5$ J/m^3, while the average distance for the correlated magnetic fluctuations is $L_0 = 48$ nm.

At intermediate temperatures, 70 < T < 205 K, the amorphous interface is ferromagnetically ordered, coupling the Fe nanoparticles by direct exchange interaction. Therefore, the system reaches a very stable long range ferromagnetic (FM) order and the global magnetization increases. By increasing the field, an increasingly stable ferromagnetic behaviour is observed.

At high temperatures (T > 210 K), the amorphous interface becomes paramagnetic, as previously observed by Chien *et al.* [5] and, therefore, the direct exchange between the nanoparticles is again disabled, and the magnetization of the system decreases. As a result, the correlation length among the

nanoparticles decreases, as corroborated by a sudden increase in the coercive field with *T*, and the system enters into superparamagnetic like (SPM) state.

Very similar magnetic behaviours have been obtained in reentrant spin glass systems, such as nanocrystalline alloys with small nanograins of Fe growth from an amorphous matrix [6]. In this kind of materials, the interactions among the Fe nanograins are highly influenced by the nature of the interface between them and the matrix.

For the Fe_xAg_{100-x} thin films with $x < 50$, both the *M(T)* and *M(H)* curves indicate a predominantly amorphous nature, even if some small Fe nanoparticles are present.

References

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Figures

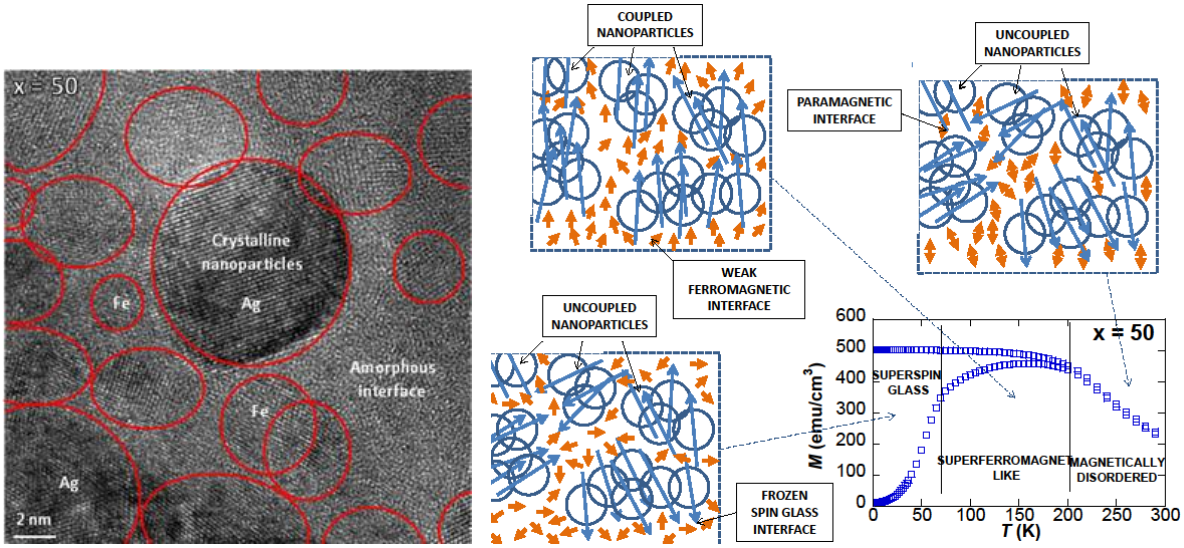

FIG1: (left) High resolution transmission electron micrograph obtained for a $Fe_{50}Ag_{50}$ sample deposited by PLD. Ellipsoids mark the positions of some crystalline nanoparticles. (right) Picture of the thermal evolution of the magnetic structure for a $Fe_{50}Ag_{50}$ thin film. Simplified pictures of the magnetic moments configurations in three ranges of temperatures are also included.