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Collective magnetic behaviors in Fe_xAg_{100-x} granular thin films ($20 < x < 50$)

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Abstract – Fe_x-Ag_{100-x} ($20<x<50$) granular thin films have been prepared by sputtering deposition. As a result, Fe nanoparticles (~ 3 nm) inside a granular Ag matrix have been obtained. At low compositions, these nanoparticles show a transition from a superparamagnetic to an spin-glass like state with decreasing temperature, mediated by dipolar interactions and their anisotropy. As the Fe concentration increases, a change in the evolution of the thermal magnetization curves (above ~ 35-39 at. % Fe) is observed, indicating a more collective magnetic behaviour due to the more important role of direct exchange interactions.

Granular materials composed of magnetic nanoparticles embedded in a nonmagnetic matrix show a rich variety of magnetic configurations that depend on the size, the concentration and the interactions among the nanoparticles. Binary Fe-Ag alloys are ideal systems to study such phenomena since Fe and Ag are highly inmiscible. In our case, we have prepared Fe-Ag thin films (~100-200 nm) in the range of 20-50 at. % Fe by sputtering deposition technique. These films were deposited at 300 K onto Si(100) substrates and coated with ~10 nm gold capping layers. Their composition was determined by energy dispersive X-ray analysis (EDX) and the microstructure was studied by X-ray diffraction (XRD) and Transmission Electron Microscopy (TEM). DC and AC zero-field-cooled/field-cooled (ZFC-FC) curves were measured using VSM and SQUID magnetometers as a function of temperature (5-350 K) and with frequencies in the range 0.03-1000 Hz.
X-Ray diffraction spectra have revealed the presence of small Fe bcc nanoparticles (~3 nm) inside a granular Ag fcc matrix (~7-10 nm). These results have been corroborated by TEM analysis.
Concerning the magnetic response, as depicted in Fig. 1 (left), the samples with lower composition show a peak in the DC ZFC curve (at T_P = 83.5K for $Fe_{20}Ag_{80}$), indicative of a blocking/freezing phenomenom at low temperatures, mediated by magnetic dipolar interactions and the anisotropy of the nanoparticles. After the peak, a Curie Weiss transition to a superparamagnetic state is observed (T_C = 117.2 K for $Fe_{20}Ag_{80}$). As the composition increases, both the peak and the transition progressively displace to higher temperatures, revealing the higher importance of direct exchange interactions. As derived from the evolution of this peak, near 35-39 at. % Fe a change in the magnetic behaviour takes place (see Fig. 1 right). From these magnetic measurements, we have estimated a size of 2.5-3 nm for our nanoparticles which apparently doesn´t change with increasing composition, suggesting that as the at. % Fe increases, so it does the number of nanoparticles, instead of their volume. In order to get a deeper insight of the magnetic behaviour in our samples, we have also performed ZFC AC magnetization measurements. As can be seen in Fig. 2, both χ' and χ'' curves show a pronounced maximum near the transition observed in DC magnetic measurements. Clear differences in the dynamical behaviour of these curves for samples below and above ~35 at. % Fe have also been observed: at low compositions the system suffers an homogeneous freezing like a superspinglass (T_g = 131.57K for $Fe_{24}Ag_{76}$) as corroborated, for example, by fitting the relaxation time to a critical slowing down law ($\tau = \tau_0(T_m/T_g-1)^{-z\nu}$, with $z\nu$ = 12 for $Fe_{24}Ag_{76}$). At higher Fe concentration, the process of dynamic relaxation of the nanoparticles becomes more collective and more similar to that of a typical phase transition ($z\nu$ = 4 for $Fe_{39}Ag_{61}$), indicating a ferromagnetic-like behaviour.

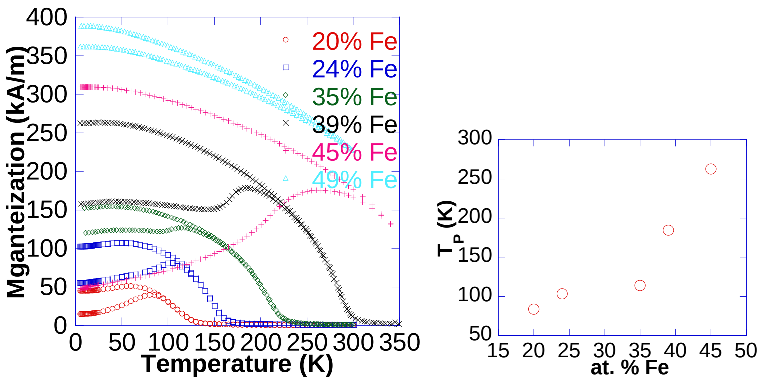

Figure 1: (left) ZFC-FC DC magnetic measurements for Fe_xAg_{100-x} ($20<x<50$) with H=5 Oe (right) evolution of the peak temperature in DC ZFC curves against the composition

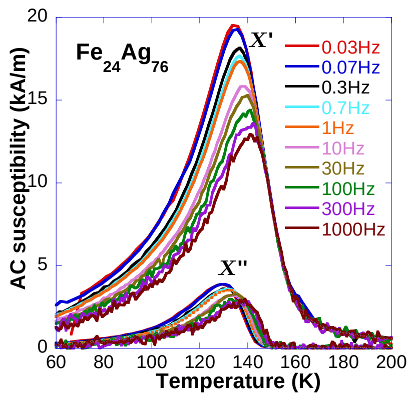

Figure 2: ZFC AC χ' and χ'' magnetic measurements for $Fe_{24}Ag_{76}$ with H=5 Oe and $0.03 < f < 1000$ Hz