## Structural, magnetic and magnetotransport characterization of Fe/MgO granular multilayers

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Magnetic granular solids consist of nanometer-sized magnetic particles embedded in an immiscible insulating or metallic medium. The spin-dependent transport in these materials is of particular interest for magnetoelectronic applications, such as sensors, read heads and nonvolatile memories<sup>1</sup>. In this work we report the structural, magnetic and magnetotransport characterization of  $[Fe(t)/MgO]_N$  multilayers with nominal Fe layer thickness *t* near percolation threshold (t~0.8 nm).

Granular multilayers  $[Fe/MgO]_N$  were prepared on glass substrates by sequential Pulsed Laser Deposition (PLD). A 3 nm buffer layer of MgO was deposited on the glass substrates. The total amount of Fe was preserved between samples by choosing the number of bilayers N, while the nominal thickness of MgO layers was fixed at 3 nm.

Specular X-ray reflectivity profiles of the multilayers show well defined first and second order Bragg peaks, and Kiessig fringes, indicating a high degree of structural periodicity of the samples [FIG.1]. TEM micrographs [FIG.2] show that the structure of each Fe layer evolves from continuous film to an ensemble of granules through multiple percolation structures with decreasing *t*.

A transition from ferromagnetic for t > 0.81 nm to superparamagnetic (SPM) for  $t \le 0.61$  nm behaviour upon decreasing iron concentration is observed. Zero Field Cooled (ZFC) and Field Cooled (FC) susceptibility measurements show clearly irreversible behaviour for SPM samples below bifurcation temperature (T<sub>b</sub>). The values of T<sub>b</sub> increase with *t* indicating growth of the average particle size.

For the films with 0.40 nm  $\leq t \leq 0.61$  nm the particle size distribution (PSD) was estimated from fitting *M vs. H* curves at different temperatures above T<sub>b</sub> using weighted Langevin functions and an approach of log-normal distribution of particle sizes [FIG.3]. The values of average particle size obtained from magnetic data fittings correlates with those from plan-view high resolution-TEM micrographs of MgO/Fe/MgO trilayers deposited on carbon grids with the same Fe:MgO ratios.

The magnetoresistance (MR=[ $\rho(H)-\rho(0)$ ]/ $\rho(0)$ , where  $\rho(0)$  and  $\rho(H)$  are the resistivity of the film in zero and in applied magnetic field, respectively) was measured with magnetic field in the films plane and parallel (L-geometry) and perpendicular (T-geometry) to the applied current in temperature range 10 – 300 K and in magnetic field up to 18 kOe. An isotropic MR ~3% at room temperature was found for the films with t  $\leq 0.61$  nm [FIG.4]. The temperature dependence of resistance for this film follows the behaviour  $\rho(0) \sim \exp(2 \cdot E_c/k_BT)^{0.5}$  indicating tunnelling type of conductance [FIG.5]. Here  $E_c$  is an activation energy necessary to create a pair of charged particles during thermally activated and/or bias assisted tunneling process in a system with distribution of particle sizes and interparticle distances,  $k_B$  is Boltzmann constant and T is temperature. Thus MR is due to spin-polarized electron tunnelling between neighboring SPM granules<sup>1</sup>. An enhanced MR is observed

[FIG.6] at low temperatures indicating higher-order tunnelling processes between large particles mediated by small ones as T decreases<sup>2</sup>. For the films with t>0.81 nm an anisotropic MR for Lgeometry measurements is observed. This confirms the formation of continuous Fe layers in the films<sup>3</sup>.



Fig. 1 X-ray reflectivity measured in the sample with nominal Fe thickness t=0.81 nm (open circles) and fitted reflectivity (solid line).



Fig. 2 Cross section TEM images of selected MgO/Fe specimens with increasing thickness (a) t=0.4 nm, (b) t=0.81 nm and (c) t=1.25 nm.

(a)



Fig. 3 Fitting of the hysteresis loops of the layered granular films Fe (0.61 nm)/MgO (3 nm) at 100 K (square), 200 K (circle) and 295 K (triangle) (left panel) and calculated particle sizes distribution (right panel).



Fig. 5 Temperature dependence of resistance for Fe(0.61nm)/MgO(3nm) film. Solid line presents results of the experimental data fitting using  $\ln(\rho) \sim (E_c/T)^{0.5}$  model.

## References



Fig. 4 Dependencies of MR on applied magnetic field for films Fe(0.61nm)/MgO(3nm) (a) and Fe(0.81nm) /MgO(3nm) (b). Measurements in L-(solid squares) and T- (open circles) geometries at room temperature.



Fig. 6 Temperature dependence of MR for Fe(0.61nm)/MgO(3nm) film in magnetic field of 18 kOe, T-geometry.

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