## Epitaxial growth of Fe<sub>3</sub>O<sub>4</sub> Thin Films and Fe<sub>3</sub>O<sub>4</sub>/MgO/Fe heteroepitaxial structures for magnetic tunnel junctions

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Magnetite (Fe<sub>3</sub>O<sub>4</sub>) is a half-metallic ferromagnet with a high Curie temperature (860 K) that is expected to exhibit suitable properties for its implementation in spintronic devices [1-3]. In addition, the interest in spintronic structures based on magnetic oxides has increased recently [4]. MgO is a serious candidate as tunnel barrier because of the small lattice mismatch with the Fe<sub>3</sub>O<sub>4</sub> (0.3%) electrode [5-7]. We believe that prior to application in real devices, the epitaxial growth of heterostructures with Fe<sub>3</sub>O<sub>4</sub> and MgO should be further optimized. Additionally, recent magnetization studies on epitaxial Fe<sub>3</sub>O<sub>4</sub> thin films grown on MgO [100] show that the ultrathin films (<5 nm thickness) are ferromagnetic and their magnetic moments are <u>much greater</u> than those of bulk magnetite, particularly at a thickness of 20 nm or below [8]. The observation of a ferromagnetic nature in ultrathin magnetite films is in contrast to the previously accepted dead layer interface model or a superparamagnetic behavior for ultrathin films of magnetite.

In this work we report the growth of epitaxial  $Fe_3O_4$  thin films and  $Fe_3O_4/MgO/Fe$  heterostructures on MgO (001) substrates by means of pulsed laser deposition (PLD) and their structural and magnetic properties.

Fe<sub>3</sub>O<sub>4</sub> and Fe thin films have been grown on single-crystal MgO (001) substrates by PLD using a KrF laser (248 nm). All layers have been deposited in ultra-high vacuum (base pressure  $< 5 \times 10^{-9}$  Torr) at substrate temperatures of 400°C for Fe<sub>3</sub>O<sub>4</sub> and between room-temperature (RT) and 400°C for Fe and MgO layers. Individual thin films and Fe<sub>3</sub>O<sub>4</sub>/MgO/Fe heterostructures on MgO (001) have been characterized by x-ray diffraction ( $\theta$ -2 $\theta$ ,  $\omega$  scans,  $\phi$  scans, and reciprocal space maps) and x-ray reflectivity (XRR), high-resolution transmission electron microscopy (HRTEM) and VSM and SQUID magnetometry. Previous magnetoresistance and anomalous Hall effect measurements in our epitaxial Fe<sub>3</sub>O<sub>4</sub> thin films have been published elsewhere [9, 10].

In Figure 1 we show the dependence of the magnetization with the nominal thickness of the films, which show a similar behavior as the observed by Arora et al [8]. For thickness below 20 nm the magnetization reaches values higher than the obtained for Fe<sub>3</sub>O<sub>4</sub> single crystals (498 emu/cm<sup>3</sup>) and for ultrathin films (< 5nm) values higher than 1000 emu/cm<sup>3</sup> can be observed. The origin of this unexpected behavior is still unclear. The non-compensation of spin moments between the tetrahedral and octahedral sublattices at the surface and antiphase-domain boundaries are inferred to be the main factor contributing to the observed enhanced magnetic moment [9].

After optimizing the growth conditions and fully characterizing the single Fe<sub>3</sub>O<sub>4</sub> and Fe layers onto MgO (001), we undertook the growth of the full Fe<sub>3</sub>O<sub>4</sub>/MgO/Fe heterostructures by PLD. HRTEM data (not shown here) demonstrate a high crystallinity of the MgO (001) tunnel barrier and sharp interfaces. From the XRR analysis in a selected heterostructure where both MgO barrier and Fe counterelectrode have been deposited at RT, a rms roughness relatively low, ~ 0.2 nm, is obtained, this being indispensable for future MTJs. The RT hysteresis loop in a sample grown in similar conditions, Fig. 2, reveals an independent switching of both Fe<sub>3</sub>O<sub>4</sub> and Fe electrodes, this also being required for tunnel magnetoresistance. The low field (500 Oe) temperature dependence of the magnetization of the same heterostructure (not shown here) displays a clear and sharp drop at the Verwey transition,  $T_V=115$  K, demonstrating the high quality of the Fe<sub>3</sub>O<sub>4</sub> layer.

We have produced high quality epitaxial  $Fe_3O_4/MgO/Fe$  heterostructures by PLD. To our knowledge, this type of heterostructure has not been grown before by PLD. Microfabrication of MTJs from these is in progress.

## **References:**

- [1] K. Gosh et al., Appl. Phys. Lett. **73** (1998), 689.
- [2] X. W. Li et al., Appl. Phys. Lett. 73 (1998), 3282.
- [3] P. Seneor et al., Appl. Phys. Lett. 74, 4017 (1999)
- [4] M. Bibes and A. Barthélémy, IEEE Trans. on Electron Devices 54, 1003 (2007)
- [5] T. Kiyomura et al., J. Appl. Phys. 88, 4768 (2000)
- [6] W. Kim et al., J. Appl. Phys. 93, 8032 (2003)
- [7] X. Jin et al, J. Magn. Magn. Mater. 286, 128 (2005)
- [8] S. K. Arora et al, Phys. Rev. B 77, 134443 (2008)
- [9] J.M. De Teresa et al, Microelectr. Eng. **84**, 1660 (2007)
- [10] A. Fernandez-Pacheco et al., submitted to Phys. Rev. B (2008)

**Figures:** 



Figure 1. Dependence of the magnetization on the nominal thickness for the  $Fe_3O_4$  thin films. The line is a visual guide.



Figure 2. Room-temperature hysteresis loop of a  $Fe_3O_4$  (70.9 nm)/MgO (3.7 nm)/Fe (5.6 nm)/Au (5.3 nm) heterostructure. The switching fields of both electrodes have been marked.