New Developments In EELS Applied To Interface Study In Magnetoresistive Tunnel Junctions With Manganites

D. Imhoff*, L. Samet* ***, M. Tencé*, A. Gloter*, F. Pailloux**, J.-L. Maurice** and C. Colliex*

* Laboratoire de Physique des Solides, Univ. Paris-Sud, F-91405 Orsay cedex France

** Unité Mixte de Physique CNRS/Thalès, F-91404 Orsay cedex, France

*** Laboratoire Physique de la Matière Condensée, FST, 1060 Tunis, Tunisia

The half-metallic ferromagnets are, in principle, ideal materials for spin electronics since they are expected to present a complete spin polarization of their electron states at the Fermi level. Record magnetoresistances (MR) have been obtained in tunnel junctions with electrodes made of half-metallic manganites, like in the La_{2/3}Sr_{1/3}MnO₃/SrTiO₃/La_{2/3}Sr_{1/3}MnO₃ system (LSMO/STO/LSMO). However, these MR effects decrease more rapidly as a function of temperature than it could be expected from the value of the Curie temperature (T_c), in the bulk materials [1-3]. The TMR of tunnel junction is essentially determined by the spin polarization in the first atomic layers of the magnetic material at the barrier/electrode interface. Therefore, the main problem in such a junction is to preserve the characteristics of the "bulk" manganite LSMO to the last atomic planes at the two interfaces with the insulating STO barrier 1.5 nm to 2 nm thick. The LSMO/STO/Co system has also been studied. To get a better insight into the structure of these interfaces and understand more clearly what are the main features affecting the spin polarization properties, we have carried out an extensive investigation of the involved interfaces down to atomic scale, using HRTEM images and EELS spectrum-images in a STEM [5]. The atomic column images show the quality and the abruptness of the pseudo-morphic growth of LSMO and STO (Fig. 1) but the contrast simulation does not allow to determine the termination of LSMO on each side of STO. Electron Energy Loss Spectroscopy (EELS), using its most recent developments has been carried out for investigating chemical composition and electronic states at apex of interfaces (environment and valence of the Mn ions ...).

This contribution presents a fine structure study (ELNES) of the absorption edges Mn-2p (Fig. 2) and O-1s, relying on other characteristic signals such as Sr atomic plasmon, La-3d, La4d, Sr-3p, Ti-2p and Co-2p edges for the relative atomic profiles across the interfaces. A summary of the characteristics and role of each interface will be presented in correlation with the measured magnetic properties. One of main results is that the valence of Mn keeps its intrinsic value of the bulk material till the last MnO₂ layers at the first interface LSMO/STO, which is consistent with the observation of magnetoresistance up to practically the Curie temperature of bulk LSMO in the junctions LSMO/STO/Co. Neither shift nor L_3/L_2 white line ratio corresponds to an enhanced Mn⁴⁺ character and an enrichment with Sr at any interface.

To carry out these local investigations, we have developed new tools of data processing to account for the large amount of spectra $(10^3 - 10^5)$ in each EELS spectrum-image. We also show the potential new methods of partial deconvolution of the instrumental functions to improve energy resolution and

multiple scattering correction as well as the correction of spatial distribution of the incident electrons. In association with the detector advances, the progress in data processing constitutes towards atomic resolved EELS spectroscopy. [5]

- [1] Y. Lu et al., Phys. Rev. B 54 (1996) 8357.
- [2] M. Viret et al., Europhys. Lett. 39 (1997) 545
- [3] J. S. Noh et al., Appl. Phys. Lett. 79, (2001) 233.
- [4] C. Jeanguillaume and C. Colliex, Ultramicroscopy 28 (1989) 252.

[5] The authors are grateful to J.P. Contour, A. Barthelemy and A. Fert of the CNRS-Thalès laboratory where these multilayers have been elaborated and studied.



FIG. 1. HRTEM picture of the LSMO/STO/LSMO tunnel junction showing the epitaxial relationship between the oxide layers. The white line schematically corresponds to a typical digital probe scan across the interface during the EELS experiments (probe step down to 0.2 nm).

FIG.2. Evolution of the Mn-2p edge across a LSMO/STO interface displayed in the second energy derivative mode. The position "0 nm" corresponds to the interface position located from elemental profile. The corresponding reference spectra for three Sr concentrations are also displayed ($La_{(1-x)}Sr_x$)MnO₃.