ESRF	**Experiment title:** **Magnetic and electronic properties of epitaxial half-metallic NiMnSb thin films**	**Experiment number:** **HE-979**
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Names and affiliations of applicants (* indicates experimentalists):

S. ANDRIEU
P. TURBAN — **Lab. Physique des Matériaux, UMR7556, Université H. Poincaré / Nancy I, Bd. Des Aiguillettes BP239,**

F. BERTRAN — **L.U.R.E. Bâtiment 209D, Centre Universitaire Paris Sud BP 34, F-91898 Orsay Cedex, France**

A. TAGLIAFERRI
C. DE NADAI
N. BROOKES — **ESRF**

Report:

In a magnetic metal/ insulator/ metal magnetic tunnel junction (MTJ), the magnetoresistive effect is related with the spin polarisation of the density of states at the Fermi level [1]. Thus the search for materials having higher spin polarisation is one of the possibilities in order to obtain the higher magnetoresistive effect in commercial devices. In the limit of a 100% polarisation the material is called an Half Metallic Ferromagnet (HFM) and the MJT device becomes an electron polariser and a spin switch. Some materials like $LaSrMnO_3$, NiMnSb, Fe_3O_4, CrO_2, seem to be good candidates for this goal since the density of states at the Fermi level is in theory close to zero for minority spins. This behaviour was only evidenced in $LaSrMnO_3$ and CrO_2 at low temperatures [2], but not in other materials up to now. NiMnSb films grown by sputtering were tested in MTJ but low magnetoresistances were always obtained [3]. All these groups came to the conclusion that the crystalline quality of the NiMnSb film and of the NiMnSb/oxide interface should be improved. In our group in Nancy, we try to clarify this point by growing and testing high crystalline quality NiMnSb films by MBE. The goal of the present experiment was to determine the polarisation at the Fermi level on such single-crystalline NiMnSb films.

The growth, structure and macroscopic magnetic properties of NiMnSb(100) films were analysed by RHEED, Auger and XPS spectroscopy, EXAFS, X-Ray diffraction, HRTEM and SQUID measurements [4]. We actually found that the NiMnSb films grow with the correct C1b structure necessary to test its possible half-metallic behaviour. The HRTEM images evidence the high crystalline quality of the films. To test directly the NiMnSb polarisation at the Fermi level, only one experiment is suitable, i.e. the spin-resolved X-Ray photoemission. Nevertheless, as the SR-XPS is sensitive to the surface, we had to solve the problem of protecting a NiMnSb film exposed to air. As previous experiments showed us that ionic bombardment definitely destroys NiMnSb films, we adopted 2 strategies:

<u>i) Measurements on the uncovered NiMnSb surface</u>

The NiMnSb films are encapsulated in a 100 nm thick Sb film covered by 5nm of V. As the Sb vapour pressure is very low, a heating up to 400C under vacuum is sufficient to eliminate Sb. V is here to protect Sb from oxidation and is firstly removed by ion bombardment.

A drastic test in order to control the preparation (i) is that **the NiMnSb surface is 4x4 reconstructed after the growth** in our MBE system. We actually get this surface reconstruction in the ID08 apparatus after ion bombardment to remove V (controlled by Auger) and annealing up to 550C. *There is consequently no doubt on the quality of the NiMnSb surface we investigated*. This is a crucial point according to other At 350C annealing, we remove almost all the capping Sb layer and we get the 1x1 surface arrangement. The dichroic signal on Mn is around 50% of the L_{III} edge and a small dichroic signal was detected on Ni. When increasing the temperature up to 450C, we still observed the 1x1 and the Mn dichroic signal increases. At 500C we started to observe a diffuse 4x4 arrangement, and the Mn dichroic signal continued to increase. Finally, at 550C, the 4x4 superstructure was clearly observed and the Mn dichroic signal reached 83% of the intensity of the L_{III} edge (fig.1). Moreover, the small dichroic signal of Ni looses the preedge feature previously present both at the L_3 and L_2 edges. This is the first observation of such behaviour. The spin moment obtained by sum rules analysis on Ni is 0.2 μ_B/at, whereas on Mn the analysis provide only a lower limit of 3.3 μB/at. This is close (within 20%) to the 3.9μB/at we determined by SQUID measurements on thick films. *To conclude, the atomic and magnetic characteristics of our NiMnSb films are in good agreement with the calculation [6].*

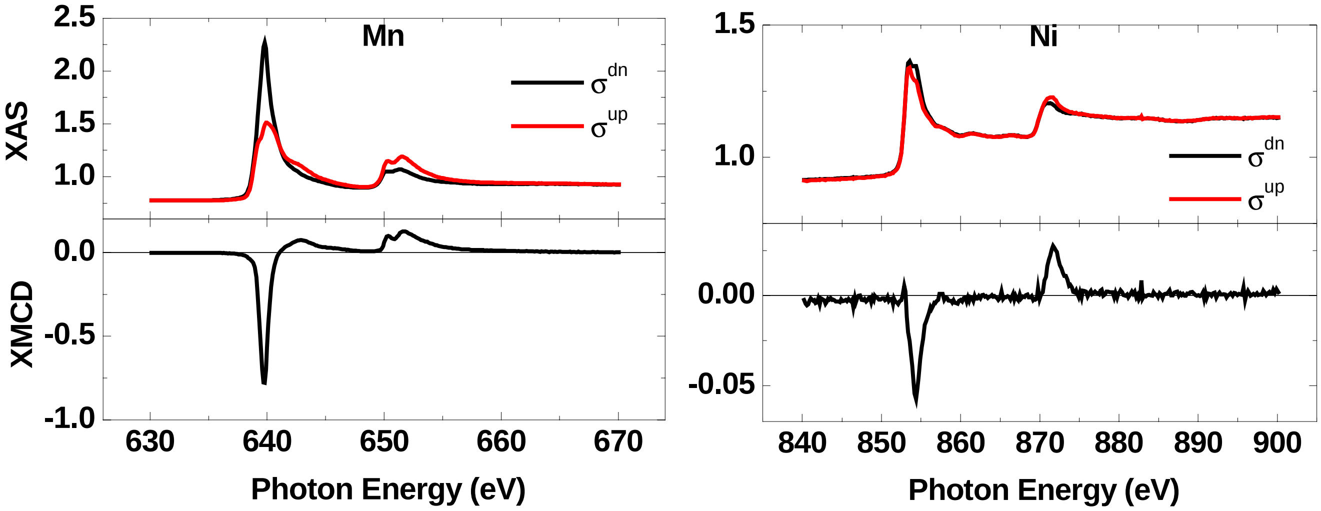

Figure 1 : absorption and dichroic spectra obtained on L_{II-III} Mn (left) and Ni (right) edges

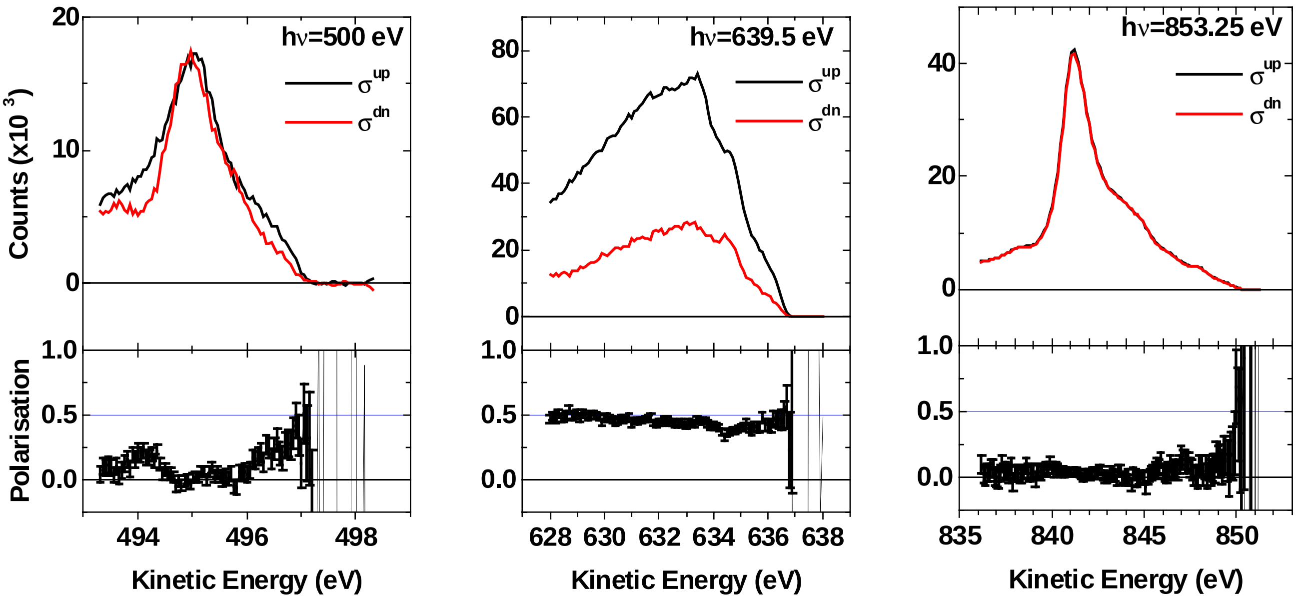

Figure 2 : SR-XPS spectra for the non-resonant (hμ=500 eV, left), resonant at Mn L_3 (hμ=639.5 eV, middle), resonant at Ni L_3 (hμ=853.25 eV, right).

The SR-XPS measurements were performed at different photon energies, i.e. at 500eV (out of resonance), at 639.5eV (L Mn edge resonance), and at 853.25eV (L Ni edge resonance). We also performed these measurements on the 1x1 and 4x4 reconstructed surface, at RT and at nearly 30K. In figure 2 are shown the spectra obtained on the 4x4 surface arrangement measured at low temperature for the three photon energies. An increase of the polarisation is clearly observed near the Fermi level up to around 40% in out-of-resonance condition. This increase is also observed in Ni edge resonance condition, and a polarisation of 50% is observed in the 10eV range investigated in Mn edge resonance condition. However, the spectra obtained in resonance condition are not easy to interpret since the dichroic contribution is also included in the measurement. Simulations are thus necessary in these cases. Finally, we did not observe significant variations of these SR-XPS spectra between low and room temperatures.

ii) Measurements on NiMnSb/MgO(100)

The NiMnSb film is protected by a thin MgO film. The goal was to measure the polarisation of NiMnSb through the gap of the covering oxide which constitutes the barrier in a MTJ. The studied sample was made of a similar NiMnSb layer than the previous one but covered by a 1.5 nm thick epitaxial MgO(100) layer. First of all, the dichroic signal obtained on Mn was not similar to those observed on the free surface: another contribution at the L_{II} edge is observed and the dichroic signal amplitude is much reduced. This is typical of some oxygen contamination since another multiplet appears in the case of MnO. We could thus estimate the quantity of oxidised Mn to be near 40%. A very important point is that this Mn-O hybridisation is not due to the NiMnSb/MgO interface since we do not obtained such spectra on NiMnSb films covered by 5nm thick MgO films measured at LURE. This may be simply explained by the fact that a 1nm thick MgO film is not thick enough to protect the NiMnSb film from oxidation in air. However, for the SR-XPS measurement, the MgO thickness must be small enough to detect some electrons coming from NiMnSb through MgO. In spite of this contamination, which may destroy the polarisation, we performed the SR-XPS measurement in order to test the counting rate through the MgO film. First, we actually detected the NiMnSb density of state in a range of 4eV below the Fermi level, in the MgO gap (fig.3). Moreover, a counting of 10 hours gave a promising statistics. We estimate that a thinner Mgo layer, protected by an additional overlayer to be removed *in situ*, should increase the counting rate enough to measure the polarisation of the NiMnSb/MgO interface by high energy spin resolved photoemission.

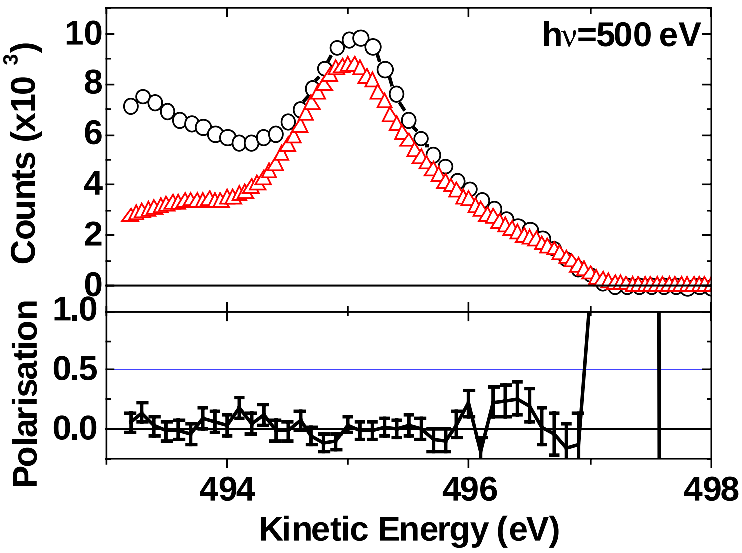

Figure 3 : Top panel: comparison of XPS spectra obtained on a NiMnSb film covered with a 1.5nm thick MgO film (circles) and free NiMnSb (triangles). Bottom panel: polarisation measured on the NiMnSb film covered with a 1.5nm thick MgO film

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