Experimental report on proposal 20130289 (32-03-718)

Elaboration of an epitaxial all-oxide magnetic tunnel junction:
growth and structure of MgO/Fe_3O_4 and $MgO/Fe_3O_4/CoO$ on Ag(001)

1/ Introduction:

This proposal aimed to study the conditions for growing an all-oxide $Fe_3O_4/MgO/Fe_3O_4/CoO/Ag(001)$ MTJ with a perfect control of the epitaxial properties. As a first step, we optimized the growing conditions of Fe_3O_4 on Ag(001), got the epitaxial parameters and roughness and looked for possible surface/interface reconstructions. In a second step, we did a $CoO/Fe_3O_4/Ag(001)$ bilayer film, elaborating then the exchange coupled counterpart of the MTJ. The $MgO(001)/Fe_3O_4(001)$ bilayer has not been done, yet. Nevertheless, one of the $CoO/Fe_3O_4/Ag(001)$ films was covered by an Al_2O_3 layer, instead of MgO, for further transport and magnetic studies.

3/ Experimental results:

In previous studies, we found the conditions for the growth of ultra-thin CoO(001) layers (~1 to 4 nm) on Ag(001) and on Fe/Ag(001) surfaces and derived their main structural parameters. We observed that, after depositing 4 ML Fe on Ag(001) and growing Co oxide on top by reactive thermal deposition, the Fe layer is completely oxidized, giving rise to a Fe_3O_4-like structure [8]. The ferromagnetic (or ferrimagnetic) character of such an interface oxide layer has been demonstrated by XMCD experiments at the Fe L-edges.

During this experimental session, the Fe_3O_4 films were grown by starting with a 4ML Fe film deposited at 300K (at 475K, in one case), with a rate of 0.2 – 0.25 ML/min, followed by oxidization at an atmosphere of $1*10^{-7}$ mbar < $p(O_2)$ < $5*10^{-7}$ mbar of oxygen. The Ag(001) substrate was cleaned using the standard UHV tools (Ar^+ sputtering, followed by annealing at about 800 K).

The structure and morphology of the films were then improved by annealing at $2*10^{-7}$ mbar of O_2 to about 650-700 K. This initial growth procedure gives rise to a well-ordered seed layer of $Fe_3O_4(001)$ with a thickness of about 1.5 – 2.0 nm. In some films, after this initial growth, the deposition was followed by Fe (4 ML) reactive thermal deposition in O_2 (Table 1), doubling the total thickness of the films.

The atomic structure of the Fe_3O_4 layer was studied using surface X-ray diffraction during growth (intermediate step and annealing) and after deposition of about a 2.0 to 3.0 nm thick films. Figure 1 shows the x-ray diffraction profile during the growth of the film.

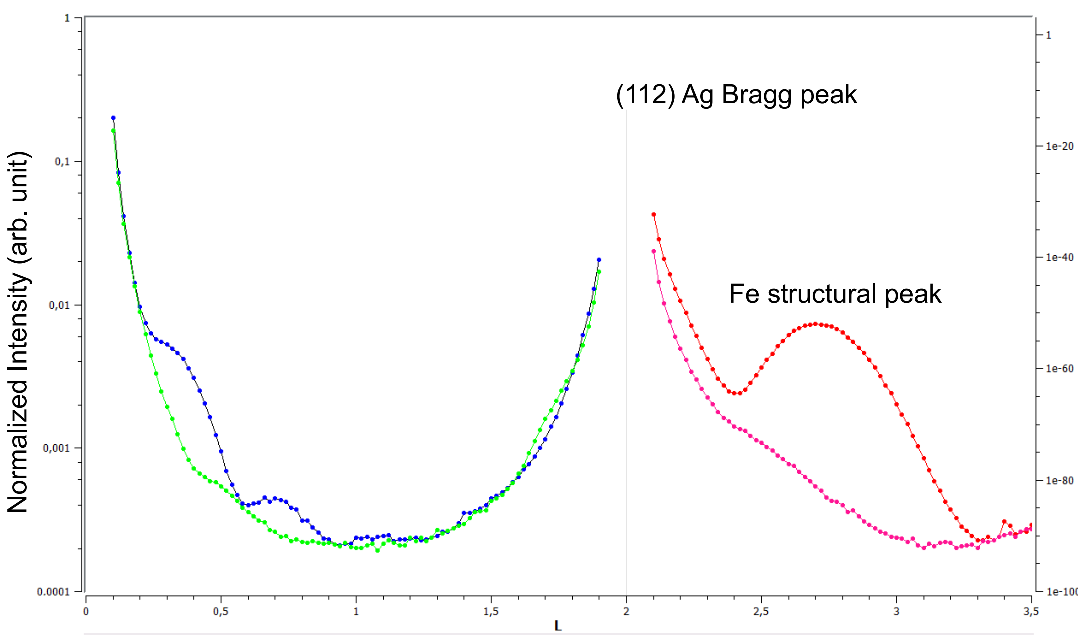

Figure 1: (1 1) CTR (crystal truncation rod) of Ag(001), after Fe deposition and after exposure to oxygen.

The surface x-ray diffraction signal after Fe deposition (blue and red curves) is observed interfering with the crystal truncation rods (CTRs) of Ag(001). This is a clear demonstration that the film is in coherent epitaxy with the substrate. A structural peak appears at L=2,7 reflecting the 4 ML Fe well-ordered with an out-of-plane parameter equal to 3,026 Å. After oxydation, the signal (green and pink curves) is very similar as the (1 1) CTR of clean Ag(001). This means that Fe is completely oxydized and it is no more pseudomorph. The diffraction intensity has been shifted to another position in the reciprocal space.

After oxydation and during the annealing, we observed the growth of the spinel (Fe_3O_4) peaks, as can be seen in the figure 2. It demonstrates the improvement of the layer order. A detailed analysis showed that these peaks correspond to the magnetite peaks.

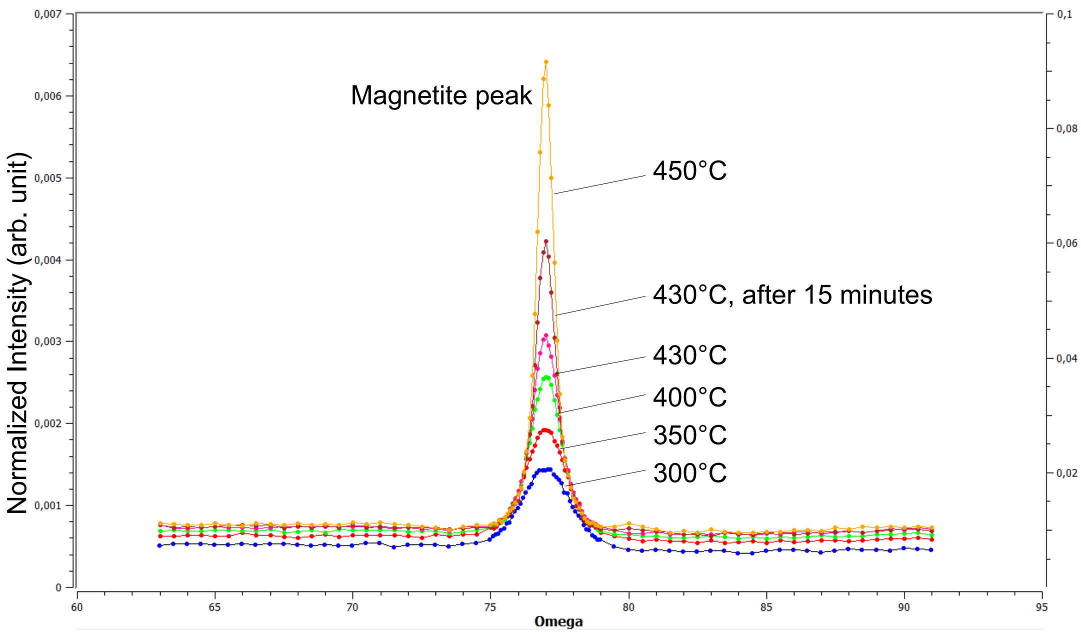

Figure 2: Magnetite XRD peak at (0,98 0,49 0,49), corresponding to a magnetite lattice constant of 8.38 Å.

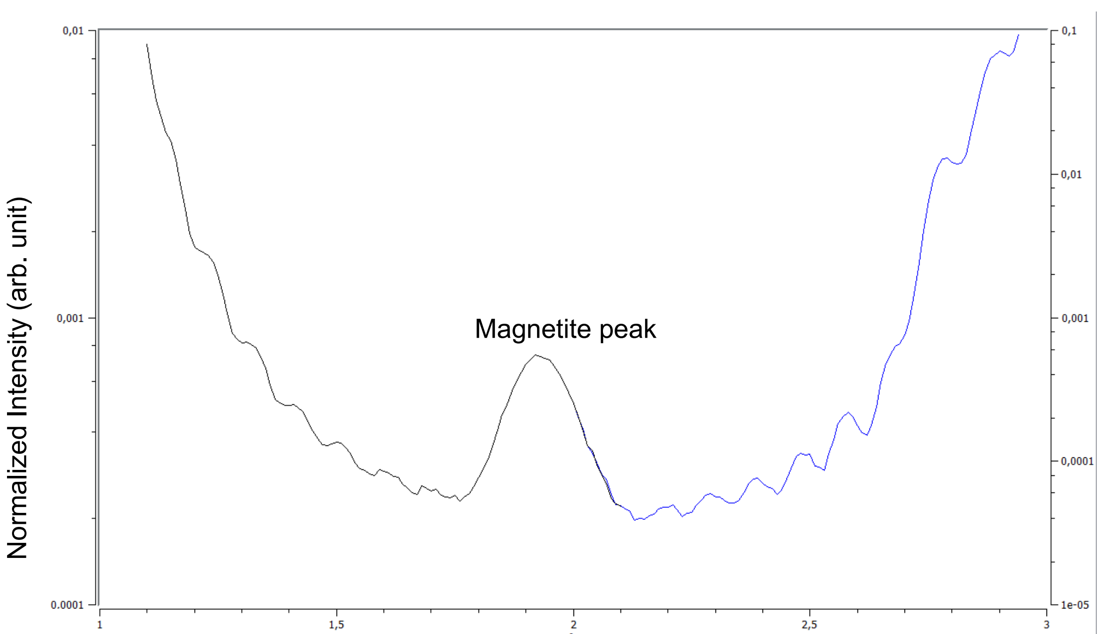

Figure 3: oxide rod (0,98 0 L) showing the magnetite peak and the oscillations associated to the well defined thickness of the final oxide layer, after CoO depostion.

In total, during this experimental session, we were able to grow five different films on different substrates. A compilation of these samples is presented at the table 1. The growing conditions were very similar, but the final structure somehow different. Two samples have 4 ML Fe and three have 8 ML Fe. One of these former are not covered by a CoO film and one has in addition a capping layer of Al2O3.

We could check that the growth of the CoO layer did not affect the structure of the Fe3O4 films. Moreover, the surface and interfaces with the substrate of the final layer is extremely flat, as indicated by the oscillations observed along the layer rods (figure 3).

	S17	S18	S20	S21	S22
Fe deposition (1)	*4 ML*	*4 ML*	*4 ML*	*4 ML*	*4 ML*
temperature	RT	475 K	RT	RT	RT
time	16'	16'	16'	16'	16'
Oxydization at RT					
pression $P(O_2)$	1x10^{-7} mbar	1.5x10^{-7} mbar	5x10^{-7} mbar	3x10^{-7} mbar	5x10^{-7} mbar
time	5'	7'	10'	10'	10'
Annealing under O_2					
pression $P(O_2)$	1.5x10^{-7} mbar	1.5x10^{-7} mbar	2x10^{-7} mbar	2x10^{-7} mbar	2x10^{-7} mbar
temperature Tmax	575 K	725 K	660 K	665 K	655 K
time at T>575 K	20'	~1h35	~35'	~30'	~20'
Fe deposition (2)			*4 ML*	*4 ML*	*4 ML*
pression $P(O_2)$			$3-5$x10^{-7} mbar	2x10^{-7} mbar	2x10^{-7} mbar
temperature			650 K - 660K	665 K	655 K
time			16'	16'	16'
Oxydization					
pression $P(O_2)$			$\sim$ 4x10^{-7} mbar	2x10^{-7} mbar	2x10^{-7} mbar
temperature			660 K	775 K	655 K
time at T>575 K			10'	1h20	14'
CoO deposition	$\approx$ *11 ML*	$\approx$ *14 ML*		*10 ML*	*6 ML*
pression $P(O_2)$	1.3x10^{-7} mbar	1.3x10^{-7} mbar		1.7x10^{-7} mbar	2x10^{-7} mbar
temperature	525 K (6.3A)	525 K		~450 K (I=5.5A)	~450 K (I=5.5A)
time	71'40"	97'		76'15"	45'
Al_2O_3 deposition					*8 Å*
temperature					Tsource=1345 K
time					16'
Oxydization					
pression $P(O_2)$					5x10^{-7} mbar
temperature					Tsource=875 K
time					5'
Comments	*Large magnetic peaks*	*Silver on surface*			*No data on Al_2O_3*

Table 1:

Table 1: it is shown the growing conditions of five different samples.

4/ Discussion:

We obtained the conditions for growing Fe_3O_4 layers of good crystallographic quality on the Ag(001) surface and extracted relevant structural parameters. The preliminary analyzes of the structure factors shows that the phase is that of the Fe_3O_4 with a good stoichiometry. We get also the conditions for growing a bilayer of Fe_3O_4/CoO, which is an important building block for the production of an echange coupled system. The MgO/Fe_3O_4/Ag(001) bilayer has not been done and additional beamtime will be necessary to complete this study. One the samples that has been produced is being prepared for transport and magnetic properties measurements.