

Experimental report on proposal:

Structural properties of perpendicularly magnetized systems: CoO/Ni and Ni/CoO bilayers grown on Pd(001)

1/ Introduction:

When a ferromagnetic (FM) thin film in contact with an antiferromagnetic (AFM) thin film is cooled through the Néel temperature of the AFM in an applied magnetic field the hysteresis loop of the FM shows enhanced coercive field and a loop-shift. This effect, called exchange bias, arises if the AFM order is established in the presence of the FM via the interfacial FM-AFM interaction. Exchange bias has been widely investigated in systems presenting in-plane anisotropy and has been applied in the magnetic recording (media) and in sensors such as spin-valves based devices, etc. Even more prominent candidates for developing new technologies are double-layer based systems combining magnetic out-of-plane anisotropy and perpendicular exchange bias effect. There is an interest also due to the fact that the phenomena of its magnetic interaction are still not yet entirely clear.

Fe-Co [1,2] and Ni [3,4] films grown on strongly mismatching fcc substrates like Pd, Ir and Rh(001) exhibit perpendicular easy magnetization axis induced by tetragonal distortion. In particular, Ni films grown on Pd(001) show perpendicular easy magnetization axis at RT, approximately up to a thickness of 15-17 ML [4]. The result confirms a volume character of perpendicular magnetic anisotropy of Ni films due to their tetragonal distortion [2,3]. Above the thickness of 17 ML of Ni the easy magnetization axis rotates towards the sample plane. A possible explanation of this change is the relaxation of the film structure which decrease the tetragonal distortion and so the perpendicular magnetic anisotropy.

Finally, it is relatively easy to grow an AFM layer, e.g., of CoO, on top of Ni(001) film. CoO seems to be a good candidate since its vertical expansion/contraction has a strong effect on electronic structure, and thus on magnetic anisotropy. Therefore, manipulation of the epitaxial strain, e.g. by growing CoO on a mismatching substrate, can change the spin axis from in-plane to out-of-plane [5]. For instance, a compressive in-plane strain (and thus the tetragonal distortion) by growing CoO on Ag(001) leads to a modified electronic structure, more precisely, to the overlap between the Co d_{xy} and oxygen orbitals different than for the bulk CoO. As a result, the orbital moment is expected to be not fully quenched, and a non-zero in-plane orbital moment is expected to exist. Thus one can expect the preferable spin orientation in the sample plane. For a similar lattice distortion (as expected for CoO film grown on top of Pd(001)), a similar effect is expected. It should be noticed, however, that the expanded in-plane lattice experienced by CoO due to its growth on MnO results in spin axis perpendicular to the sample plane. It should be mentioned, that also preparation condition of CoO can influence the growth orientation [6] which may have a strong impact on the magnetic properties.

Our recent MOKE study shows that when CoO layer is deposited on Ni film on Pd(001) (which has in-plane magnetic anisotropy), it changes remarkably magnetic anisotropy of the system [4]. At temperatures lower than Neel temperature of CoO ($T_{N,CoO}=293K$), the CoO(001)/Ni bilayers show perpendicular magnetization and exchange bias effect up to the thickness of Ni much larger than the thickness up to which Ni films on Pd(001) show perpendicular anisotropy. We applied a simple method to prove whether the temperature at which the perpendicular anisotropy starts to decrease is related to the antiferromagnetism of CoO. We oxidized the topmost 1-2 atomic layers of Ni film and then covered with another 3ML of CoO (this results in the Neel temperature remarkably increased since $T_{N,NiO}=525K$). The procedure causes perpendicular magnetization to be restored in the same thickness range but even at RT, which confirms the effect to be due the coupling to the antiferromagnetic film on top of Ni.

2/ Results:

The aim was to study the growth and the structural properties of both CoO and Ni on Pd(001) and then how the second layer stack on first one, i.e., how CoO grows on Ni/Pd(001) and Ni grows on CoO/Pd(001).

We used in situ grazing incidence x-ray diffraction (GI-XRD) at the French CRG BM32 beamline at the European Synchrotron Radiation Facility (ESRF, France) to study and optimize the growth of ultrathin Ni and CoO films on a clean Pd(001) substrate. The substrate was prepared by Ar sputtering

and annealing cycles. The Ni layer was grown by thermal deposition at a rate of 0.25ML/min at a base pressure of 1×10^{-10} mbar on the clean Pd(001) substrate held at 300 K. The layer grows at coherent epitaxy and displays a large tetragonal distortion up to about 6 ML. After that, it starts to relax the in-plane strain. From 15 to 23 ML it is almost completely relaxed with the epitaxial relation Ni(100)/Pd(100). The thickness measured by the period of the Kiessig oscillations in the final layer was found to be 22 ML, very close to the nominal one (23 ML) given by the quartz balance calibration. The lattice constant parallel and perpendicular to the surface are 3.59(2) Å and 3.47(2) Å, respectively, resulting in a $c/a=0.97(1)$.

The CoO layer was grown by reactive thermal deposition on the ultrathin (22 ML)-Ni(001) layer held at 375 K. First of all, 1 ML Co was deposited on top of the Ni film under ultra-high vacuum (2×10^{-10} mbar). The increase of the Ni truncation rods intensity and the decreasing of the peak width indicated that the Co layer is pseudomorph on the Ni(001) surface. Exposure to a partial oxygen pressure (8×10^{-7} mbar during 5 minutes) leads to the formation of an epitaxial (001) oriented rocksalt-like film with in-plane lattice constant of 4.15(2) Å. Its average thickness is estimated by the peak width perpendicular to the surface (perpendicular momentum transfer) and roughly corresponds to about 4 mixed atomic monolayers. However, the out-of-plane lattice parameter could not be measured precisely at this step. Both a decrease of the intensity of the metallic (Ni) rods and a widening of their peak were observed. This widening corresponds to a thickness reduction of 2 ML (1 Co + 1 Ni). Further deposition of 2 ML Co under a partial oxygen pressure (8×10^{-7} mbar) results in an increase of the oxide layer thickness, while the Ni film remains unchanged. The lattice constant parallel and perpendicular to the surface are 4.18(2) Å and 4.31(1), respectively, indicating a slightly in-plane compressed oxide layer with c/a of about 1.03. The Ni lattice constants did not change during the whole oxide growth process. The average Co(Ni)O oxide layer thickness has been estimated to be 1.3 nm, i.e. about three CoO lattice parameters.

Reactive thermal deposition of CoO on pure Ni(001) oxidizes then about 1-2 ML of Ni. The interface between the Ni(001) and CoO layers shows a small Ni oxide contribution related to Ni atoms dispersed within the CoO layer and/or from Ni-O bonds at the interface. Such a Ni oxide component leads to the small increase in the Néel temperature of the $\text{Co}(1-x)\text{Ni}(x)\text{O}$ layer observed by MOKE and XMLD studies. We show that the ferromagnetic properties are characteristic of metallic Ni and are not affected by the small oxide contribution. A short letter is being prepared to present the structural results along with the magnetic properties [4] (see attached draft). The detailed growth procedure and x-ray diffraction study of CoO on both Ni(001) and Pd(001) will be presented elsewhere.

6/ References:

- [1] "Strongly enhanced orbital moment by reduced lattice symmetry and varying composition of $\text{Fe}_{1-x}\text{Co}_x$ alloy films", F. Yildiz, F. Luo, C. Tieg, R. Abrudan, X. L. Fu, A. Winkelmann, M. Przybylski, J. Kirschner, *Phys. Rev. Lett.* **100**, 037205 (2008)
- [2] "Volume contribution to perpendicular anisotropy in $\text{Fe}_{0.5}\text{Co}_{0.5}$ alloy films on Pd(001), Ir(001), and Rh(001)", F. Yildiz, M. Przybylski, J. Kirschner, *Journal of Applied Physics* **105**, (7), pp 07E129/1-3 (2009)
- [3] "Magnetic anisotropy and magnetostriction in tetragonal and cubic Ni", O. Hjortstam, K. Baberschke, J.M. Wills, B. Johansson, O. Eriksson, *Phys. Rev. B* **55**, (1997) 15026.
- [4] "Effect of CoO/Ni exchange coupling on perpendicular magnetization of Ni films on Pd(001)", P. Kuswik, P. L. Gastelois, H.C.N. Tolentino, M. De Santis, A.Y. Ramos, M.M. Ramos, M. Przybylski, J. Kirschner, *Phys. Rev.*, submitted
- [5] "Controlling Orbital Moment and Spin Orientation in CoO Layers by Strain", S. I. Csiszar, M. W. Haverkort, Z. Hu, A. Tanaka, H. H. Hsieh, H.-J. Lin, C. T. Chen, T. Hibma, L. H. Tjeng, *Phys. Rev. Lett.* **95**, 187205 (2005).
- [6] "Tuning the Growth Orientation of Epitaxial Films by Interface Chemistry", M. Gubo, Ch. Ebensperger, W. Meyer, L. Hammer, K. Heinz, F. Mittendorfer, J. Redinger, *Phys. Rev. Lett.* **108**, 066101 (2012).

7/ Team :

Institut Néel, Grenoble: Maurizio De Santis, Hélio Tolentino, Anne Lamirand, Aline Ramos.

Max Plank Institut, Halle, Germany: Marek Przybylski, Piotr Kuswik.