Thin films and multilayers for spintronics deposited by a CVD-ALD process

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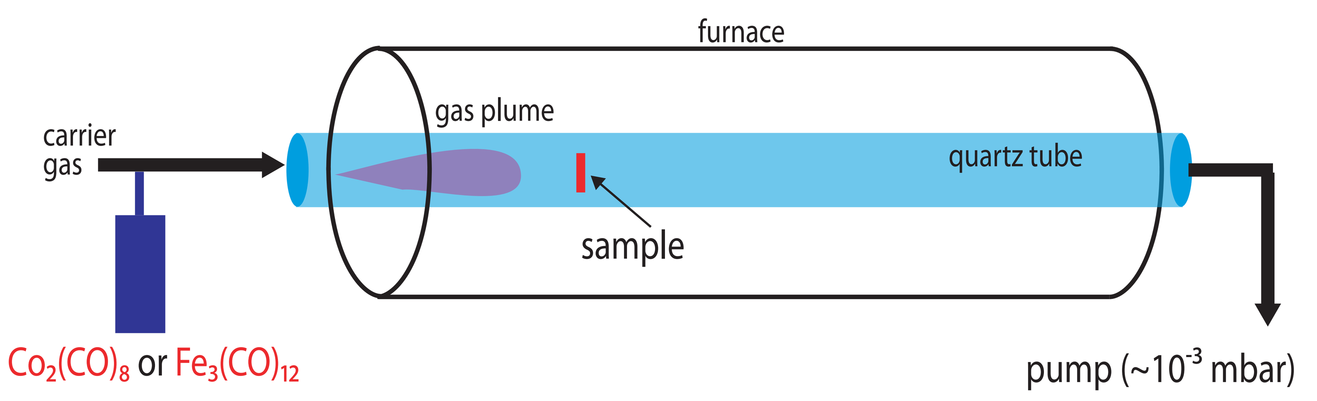
Abstract

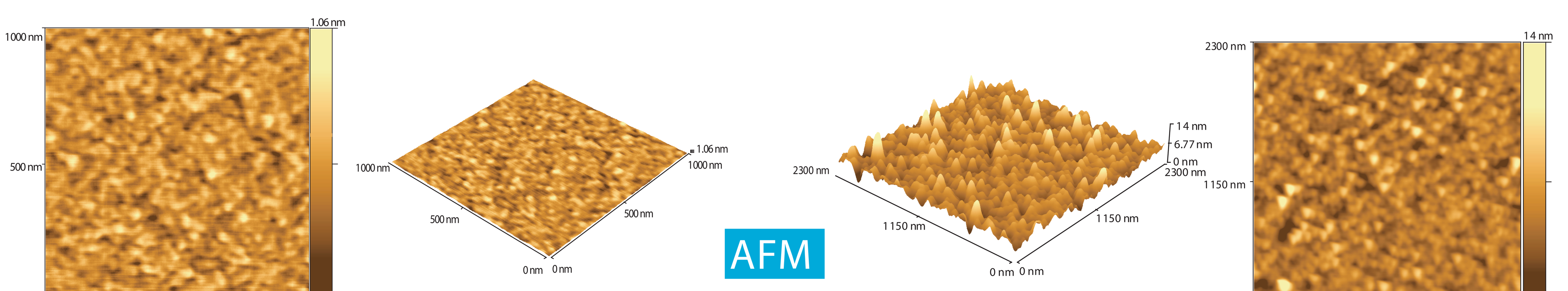
Spin-based electronics (spintronics) is receiving significative interest, as it offers possible attractive solutions for the future information technology market [1]. Among the most competitive options for replacing and/or integrating the currently used non-volatile memories, the magnetoresistive random access memory (MRAM) is very promising. The core element in MRAMs is the magnetic tunnel junction (MTJ) [1]. Typically a MTJ consists of two ferromagnetic (FM) layers acting as electrodes separated by an oxide tunnel barrier: the magnetization of the soft electrode consitutes the storage information. Reading process is achieved through the sensing of two different perpendicular-to-plane resistances in the MTJ stack, depending on the relative orientation of the electrodes magnetization, parallel or anti-parallel configurations (tunnel magnetoresistance effect). The interest that major companies are manifesting in MRAMs motivates the development and optimization of thin films deposition methods capable of growing smooth, uniform and conformal FM layers and oxides for their inclusion into functional spintronic devices. We show our recent research efforts towards the use of atomic layer- and chemical vapour- deposition (ALD-CVD) methods for the fabrication of MTJs. We developed a simple, efficient, and cost effective deposition chamber for the growth of Co and magnetite (Fe_3O_4), by employing $Co_2(CO)_8$ and $Fe_3(CO)_{12}$ carbonyls precursors, while a conventional ALD reactor was used for the deposition of thin MgO films, in such a way that the fabrication of Fe_3O_4/MgO/Co stacks was achieved. We investigated the structure, microstructure, morphology, chemical profiling, contaminations and magnetic behaviour of selected Co, Fe_3O_4, MgO films and Fe_3O_4/MgO/Co multilayers, by means of time of flight secondary ion mass spectroscopy (ToF SIMS), X-ray diffraction (XRD), X-ray reflectivity (XRR), conversion electron Mössbauer spectroscopy (CEMS), and superconducting quantum interference device (SQUID) magnetometry. The magnetoresistance (MR) of Fe_3O_4 films reaches -2.4 % with an applied field of ±1.1 T, being in accordance with the MR observed for magnetite films produced by sputtering [2].

[1] C. Chappert, A. Fert, and F. N. Van Dau, Nature **6**, 813 (2007) - [2] J. M. Coey, A. E. Berkowitz, Ll. Balcells, F. F. Putris, and F. T. Parker, APL **72**, 734 (1998)

Experimental



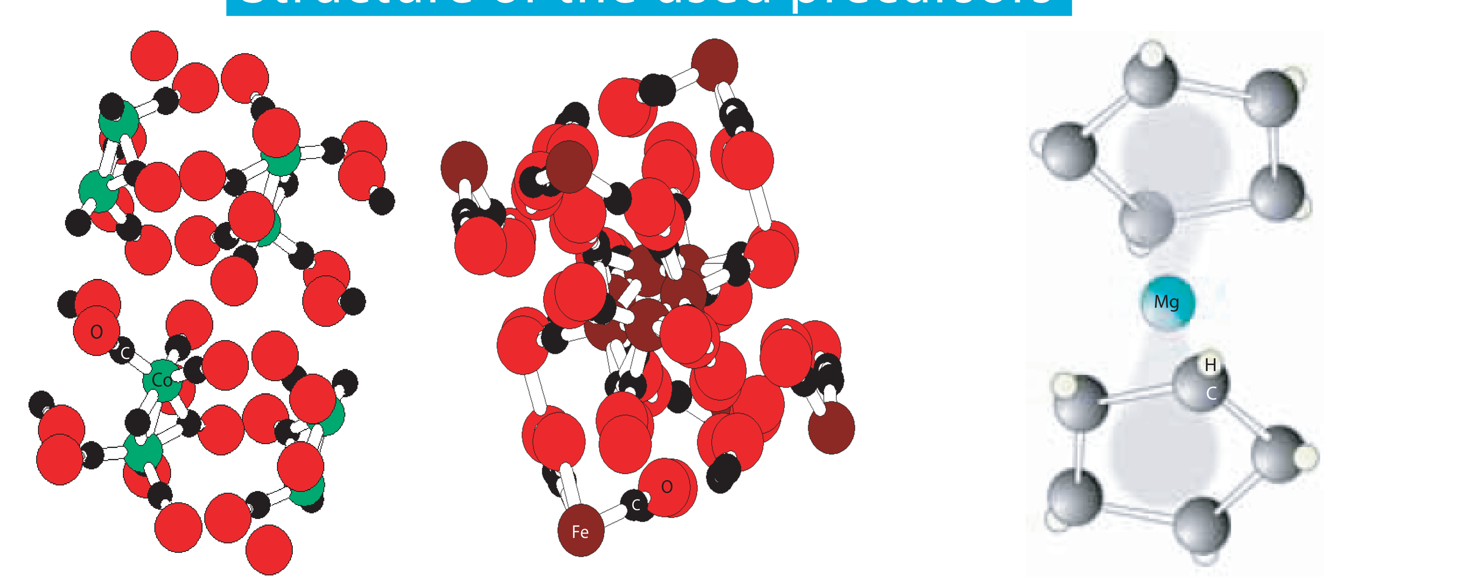
Schematic illustration of the CVD-ALD deposition sytem for the growth of Co and Fe_3O_4 films. The tunnel oxide (MgO) is grown in a conventional ALD reactor by using the $Mg(C_2H_2)_5$ precursor and H_2O as oxygen source.



MgO (5 nm thickness) r.m.s roughness 0.1 nm

Co (50 nm thickness) r.m.s roughness 1.3 nm

Structure of the used precursors



bis-octacarbonyl-cobalt $Co_2(CO)_8$ → CVD → Co

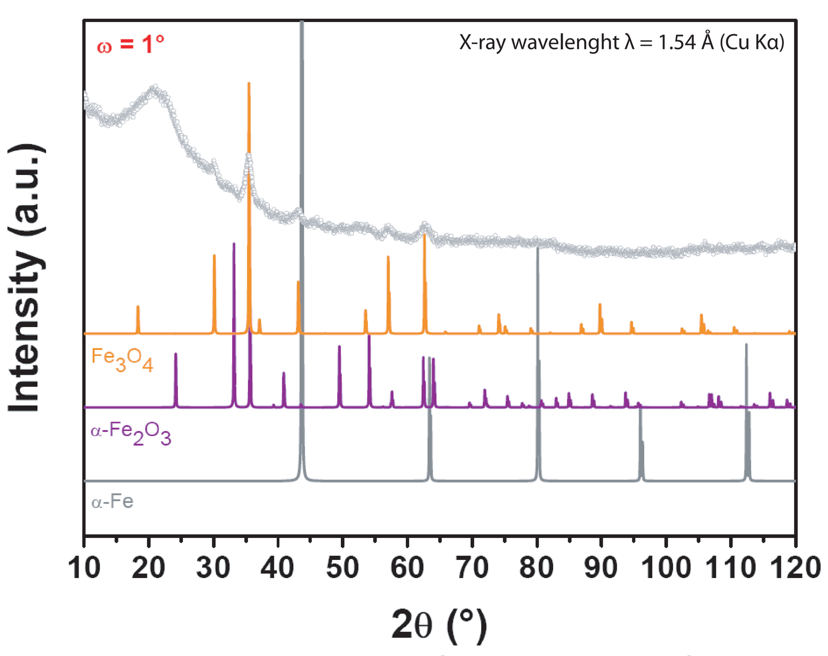
tri-dodecarbonyl-iron $Fe_3(CO)_{12}$ → CVD → Fe_3O_4

bis-cyclopentadienyl-magnesium $Mg(C_5H_5)_2$ + H_2O → ALD → MgO

Magnetite - Fe_3O_4

-half metallicity predicted
-Curie temperature 860 K

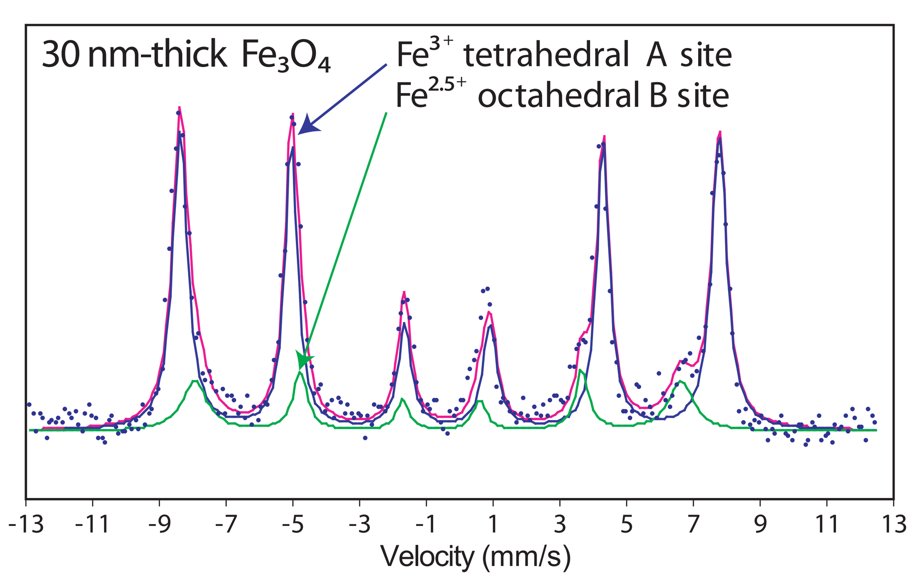
GIXRD



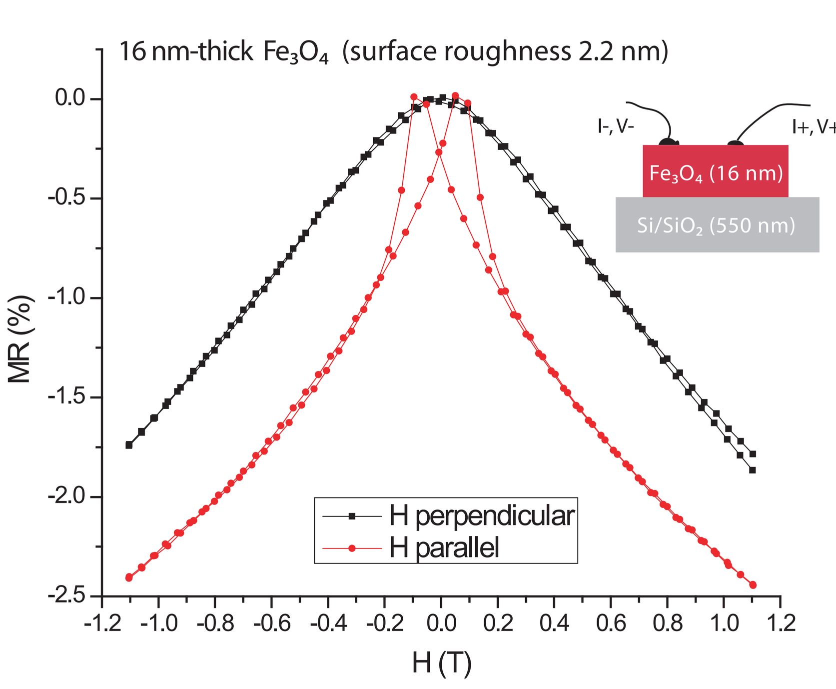
GIXRD shows the formation of poly-crystalline Fe_3O_4 (phase FD3-MZ)

CEMS

CEMS evidences the presence of two characteristic magnetically-split sextets related to the tetrahedral A site and the octahedral B site, characteristic of bulk magnetite. The Fe_3O_4 stoichiometry is deduced from the relative intensities of the sextets [3]: $(Fe^{3+})_A(Fe_{0.8}^{2.5+}Fe_{1.0}^{3+}\square_{0.2})_B O_4$

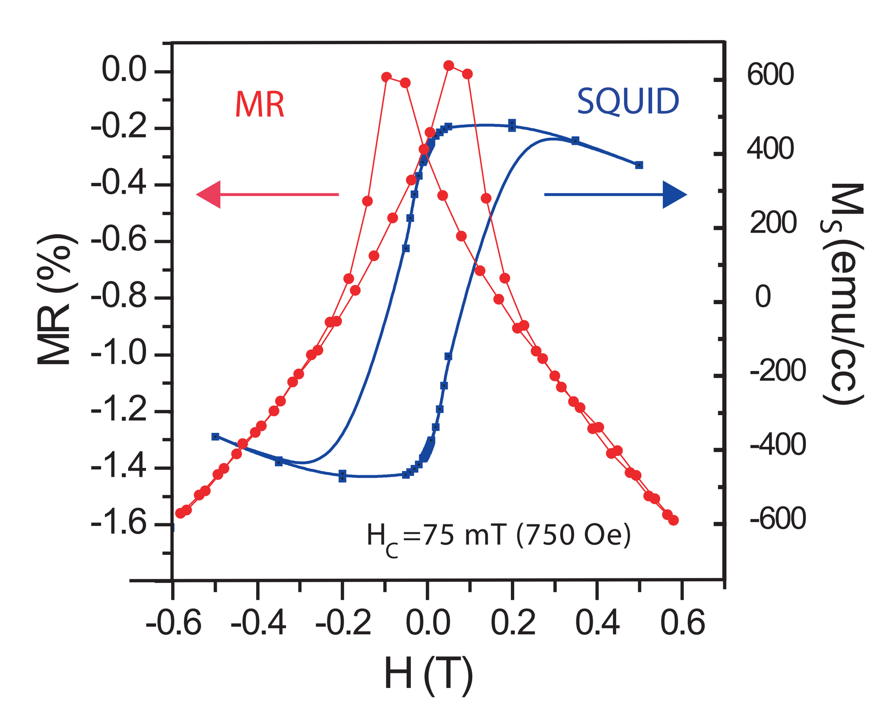


MR



For intergranular tunneling of the spin polarized electrons, the spin polarization can be deduced from the MR data [4]. Our Fe_3O_4 films show P~16%.

MR - SQUID



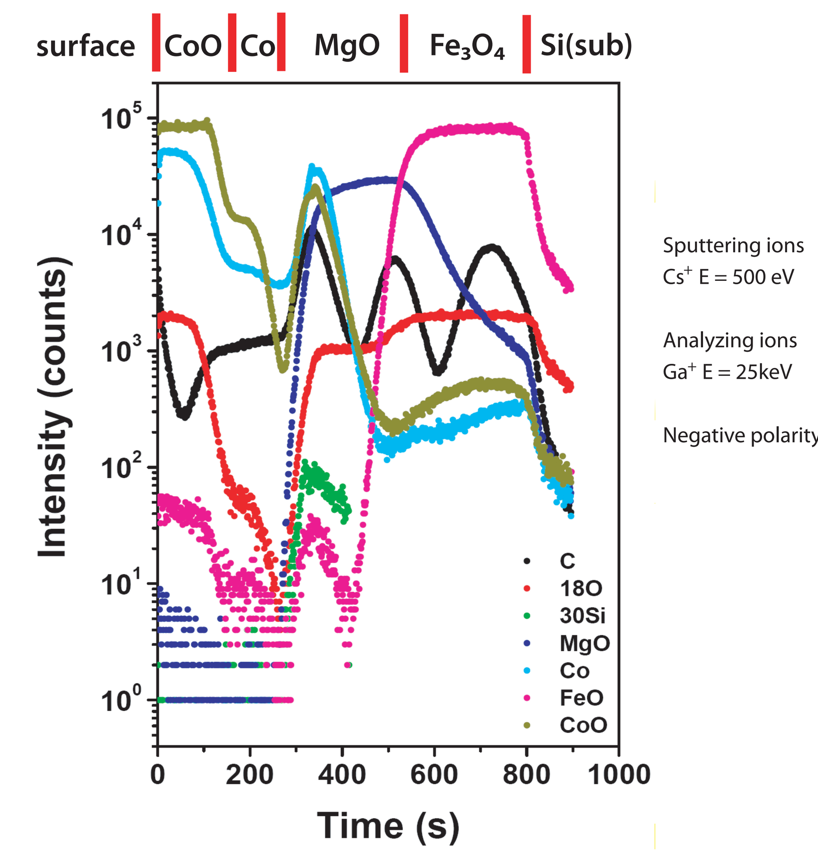
[3] F. C. Vogt *et al.*, Surface Science **331**, 1508 (1995)
[4] J. Inoue and S. Maekawa, PRB **53**, R11927 (1996)

- Pure magnetite films (XRD)
- Stoichiometry (CEMS)
- Large coercivity (SQUID)
- MR up to -2.4 % at ±1.1 T
- Spin polarization P~16%

Fe_3O_4/MgO/Co stack by CVD-ALD

ToF SIMS

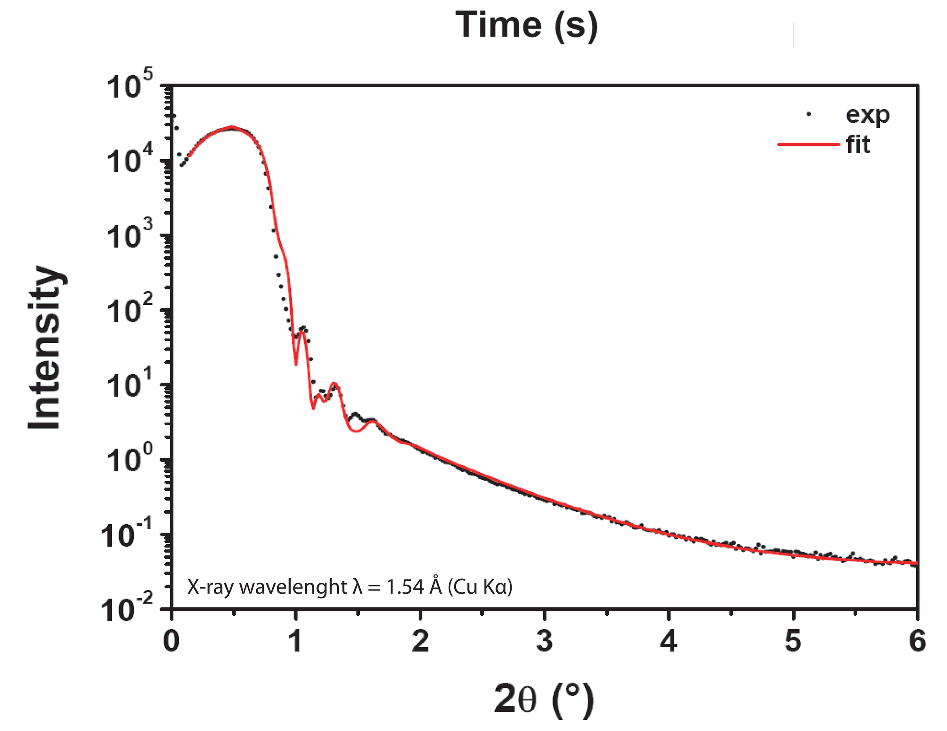
ToF SIMS depth profile of a typical Fe_3O_4/MgO/Co stack as produced with our CVD-ALD process. Co, MgO, and Fe_3O_4 layers are well distinct in the stack. Each layer is uniform and homogeneous as shown by the profile flatness. At the surface/sub-surface region Co is oxidized. No relevant contributions from precursors volatile components (such as C shown in figure) is revealed.



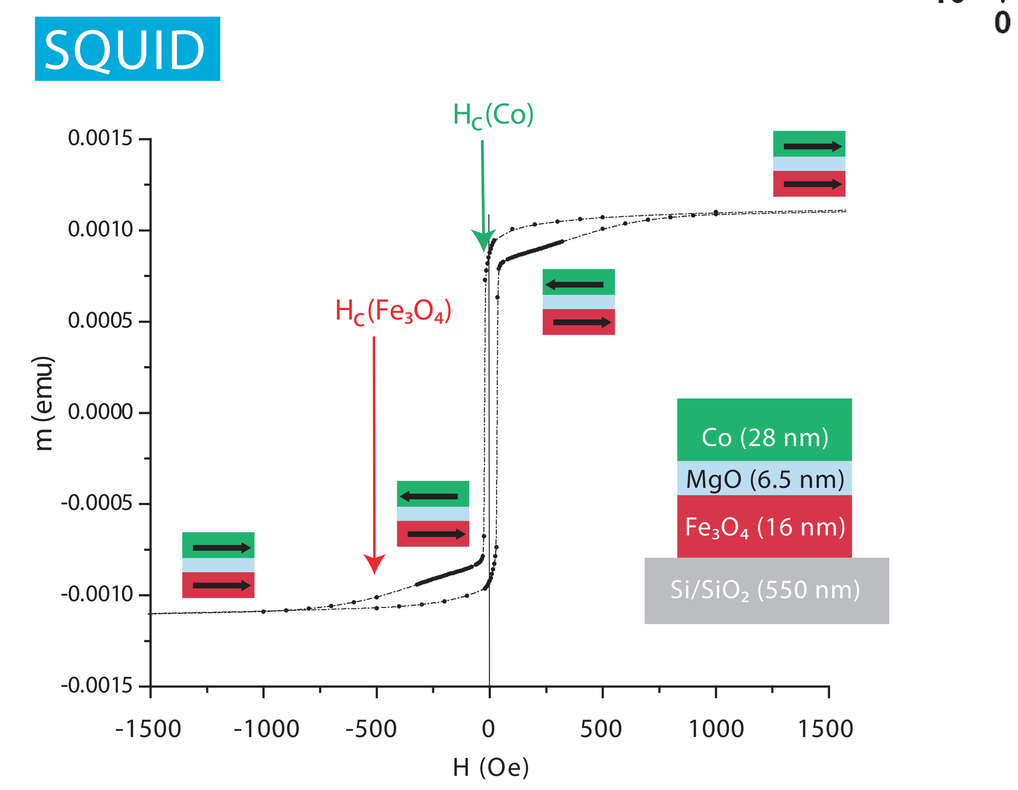
XRR

XRR measures MgO thickness of 6.5 nm with interface roughness of 2.4 nm (Co/MgO) and 3.9 nm (MgO/Fe_3O_4). Co top surface is 3.4 rough.

Layer	Thickness (nm)[±0.1]	Roughness (nm)[±0.1]	El.density ($e^- Å^{-3}$)[±0.05]
Co	28.0	3.4	2.26
MgO	6.5	2.4	1.17
Fe_3O_4	16.1	3.9	1.44
SiO_2	∞(550nm)	0.4	0.807



SQUID



The theoretically predicted half-metallicity of magnetite, the notable difference in the coercivity values of the Co and Fe_3O_4 ferromagnetic electrodes and the overall good quality of the as deposited films and multilayers, suggest that

Fe_3O_4/MgO/Co

produced by the ALD-CVD method, is promising for the fabrication of well-performing MTJs.

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