Influence of the growth conditions on the magnetism of $SrFe_{12}O_{19}$ thin films and the behavior of $Co/SrFe_{12}O_{19}$ bilayers

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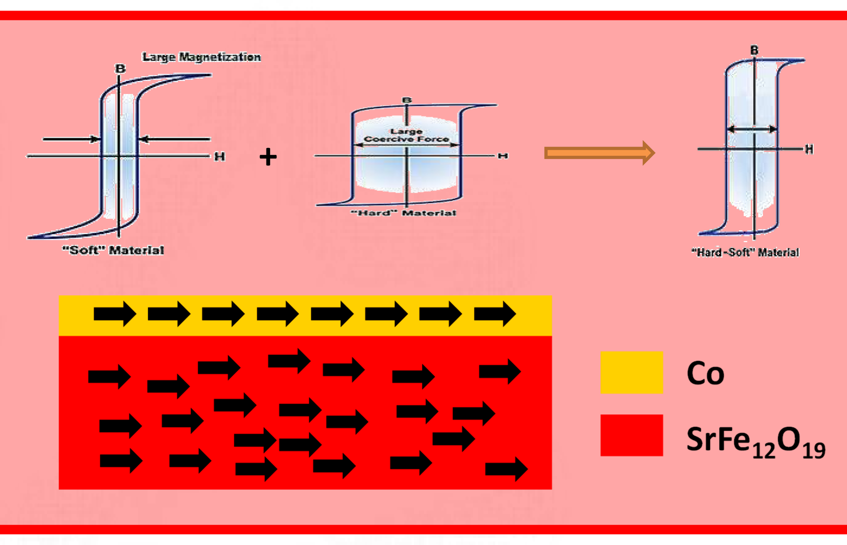
INTRODUCTION

Permanent magnets are used in a wide variety of applications, such as generators and motors in the automotive industry, household appliances, etc. Many of these magnets contain rare earths, which are critical elements whose extraction is harmful to the environment and which present a risk of price volatility. Therefore, their substitution by cheaper and environmentally friendly materials is sought. In our case, we have focused on hard strontium hexaferrite $SrFe_{12}O_{19}$ (SFO) as a basis for alternative permanent magnets. However, its saturation magnetization is moderate, M_s = 92-74 $Am^2\ Kg^{-1}$. It is well known that, rigid exchange coupling between a magnetically hard and a soft material can lead to higher remanent magnetization and avoid high coercivity loss. As a consequence, the energy product of the $(BH)_{max}$ system can be improved.

In previous studies, we examined the magnetic coupling between SFO platelets with out-of-plane magnetization and a cobalt layer [1]. It was observed that there was no correlation between the magnetic domains of the Co layer and SFO platelets, due to the absence of exchange coupling at the interface and competition of the shape anisotropy of the metal layer with the magnetodipolar field created by the SFO layer. To avoid the competition with the shape anisotropy of the layer, we have devised here an experiment using in-plane magnetized SFO films [2].



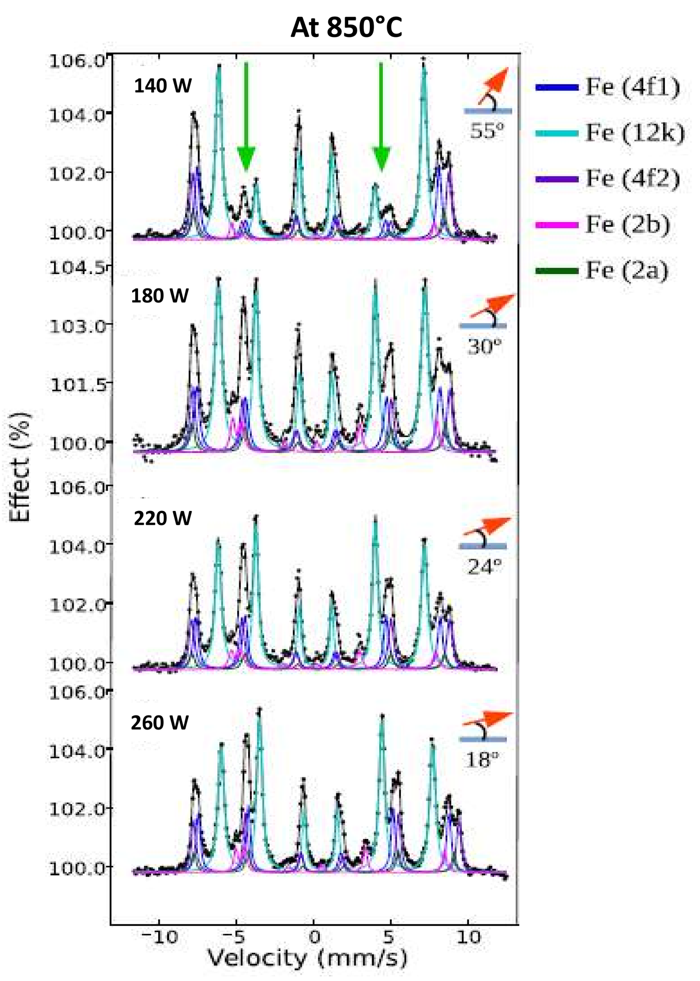
EXPERIMENTAL

<u>Experimental growth parameters by RF Magnetron Sputtering:</u>

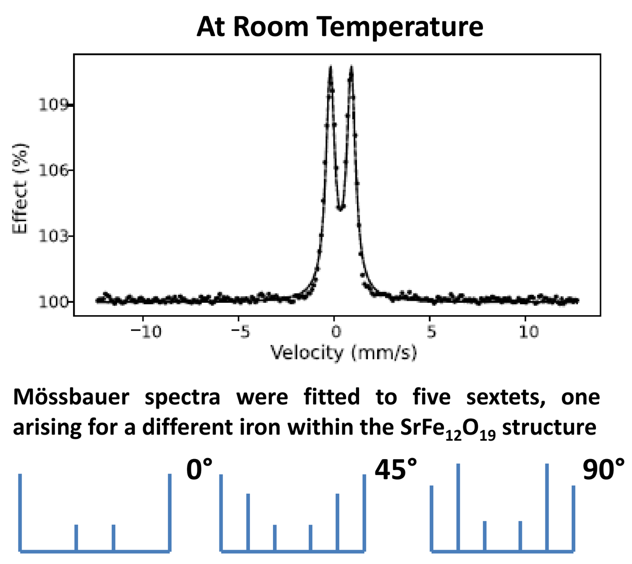
- Initial pressure ➔ 10^{-6} mbar
- Rate pressure ➔ Ar/O_2 (2%)
- Distance Substrate/Target ➔ 60 mm
- Work pressure ➔ $7\cdot10^{-3}$ mbar
- Time of pre-sputtering ➔ 15 min
- Time of sputtering ➔ 3 hours
- Annealing ➔ at 850°C during 3 hours

<u>Substrate</u>: Si [100] waffers
<u>Target</u>: commercial $SrFe_{12}O_{19}$

MÖSSBAUER SPECTROSCOPY



Mössbauer spectra were fitted to five sextets, one arising for a different iron within the $SrFe_{12}O_{19}$ structure

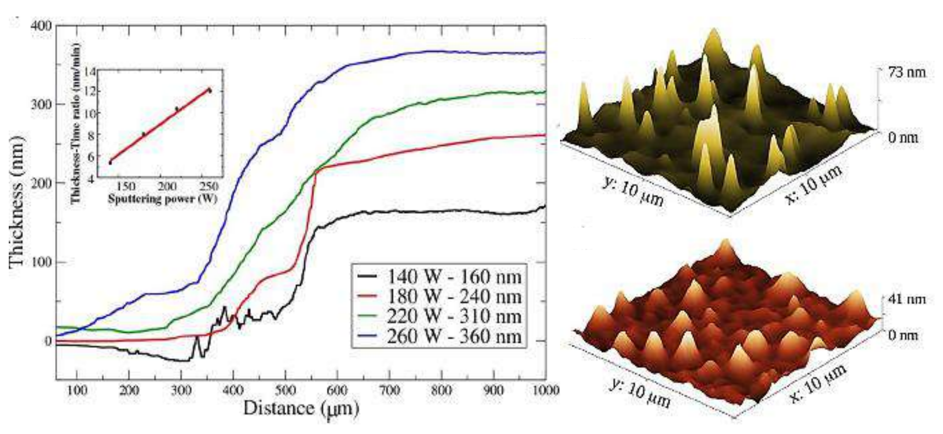


The intensities in the peaks of each spectrum allow us to estimate the orientation of the magnetization of each simple respect to the sample plane

For samples grown at 140 W, we obtain a magnetic orientation almost outside the plane of the $SrFe_{12}O_{19}$ film. As we increase the power, we observe that the magnetization tilts towards the film plane, and with 260 W is completely in-plane.
One of the reasons that could explain this magnetic change would be that at intermediate power some areas grow in the Z axis while a higher powers the film expands and grows more uniform.

CHARACTERIZATION AND RESULTS

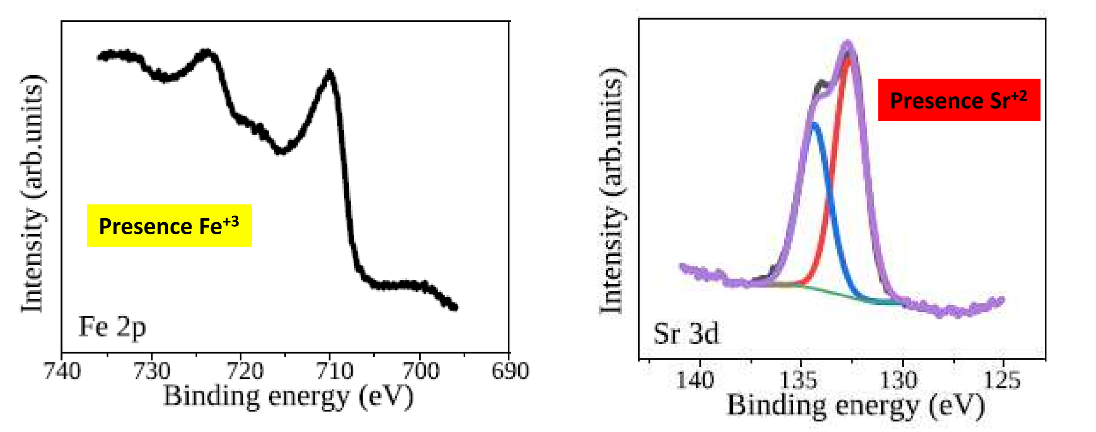
AFM



Strontium hexaferrite samples were grown by RF Magnetron sputtering at different sputtering powers (140 W, 180 W, 220 W and 260 W) for 30 minutes. By measuring the step between the thin film and the substrate the thin film thickness

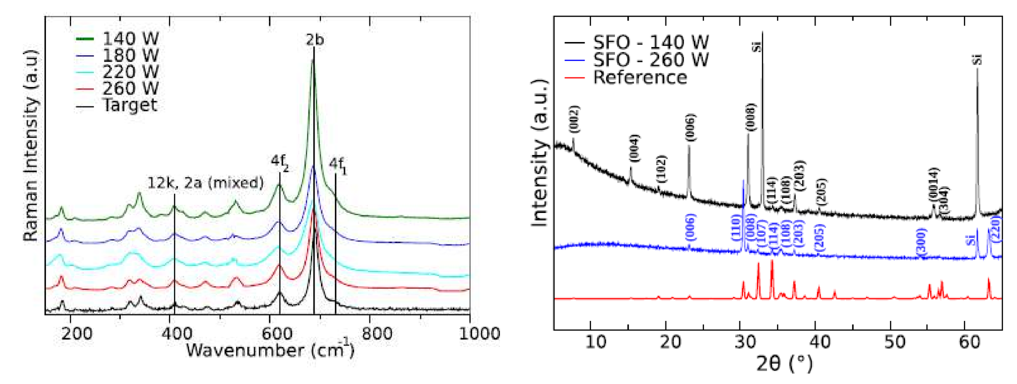
At intermediate powers ➔ columnar growth
At high powers ➔ flatter and denser surfaces

XPS



- **spin orbit-doublet (710,2 eV and 723,3 eV)**
- **small shake-up satellite at 718.1 eV**
- **spin orbit-doublet (132,6 eV and 134,4 eV)**

XDR and RAMAN



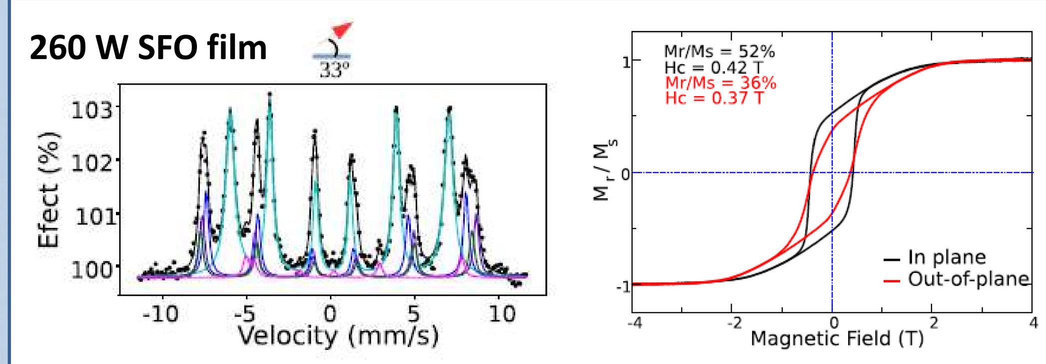
The spectra include bands at 410, 620, 688 and 734 cm^{-1} are assigned to the vibrational modes arising from the different Fe^{3+} chemistries and that are characteristic of strontium hexaferrite.
The commercial strontium ferrite target confirms that the grown films have indeed the composition of SFO

At 140W ➔ characteristic peaks of lower intensity ➔ suggesting a preferential c-axis orientation normal to the film

At 260W ➔ characteristic peaks of higher intensity ➔ c-axis whitin the plane

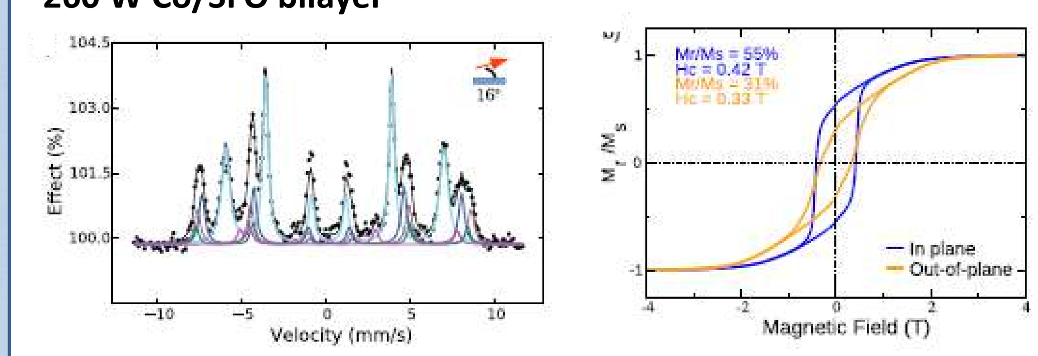
MAGNETIC PROPERTIES

260 W SFO film



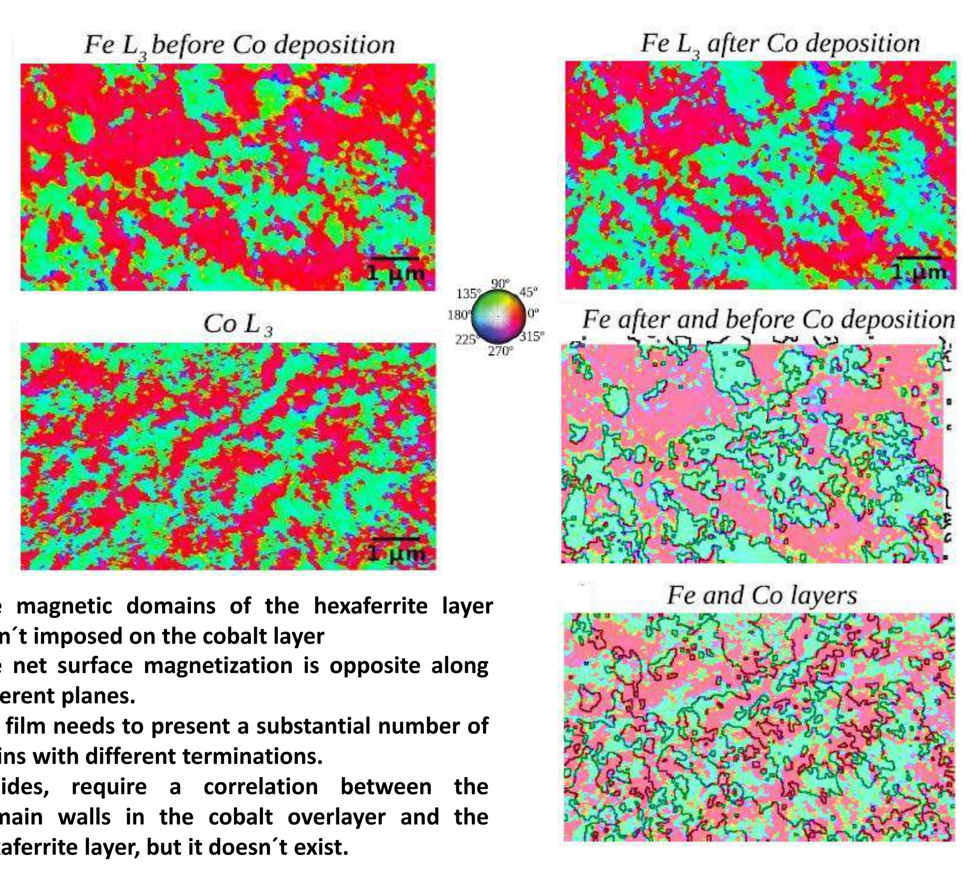
It is confirmed that the SFO film has an easy magnetic axis mainly in the plane of the sample

260 W Co/SFO bilayer



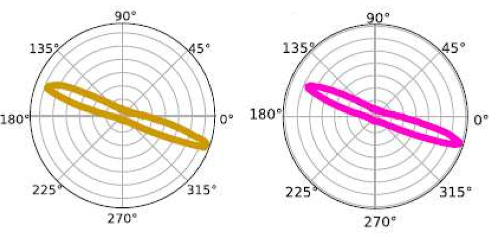
It is confirmed that the magnetization is still preferentially in-plane after Co deposition (thickness 2 nm)

XMCD



The magnetic domains of the hexaferrite layer aren´t imposed on the cobalt layer
The net surface magnetization is opposite along different planes.
the film needs to present a substantial number of grains with different terminations.
Besides, require a correlation between the domain walls in the cobalt overlayer and the hexaferrite layer, but it doesn´t exist.

The most prominent result is that the SFO film shows a uniaxial anisotropy large enough as to show a magnetization vector preferentially aligned in one direction within the film plane (160° and 340°)

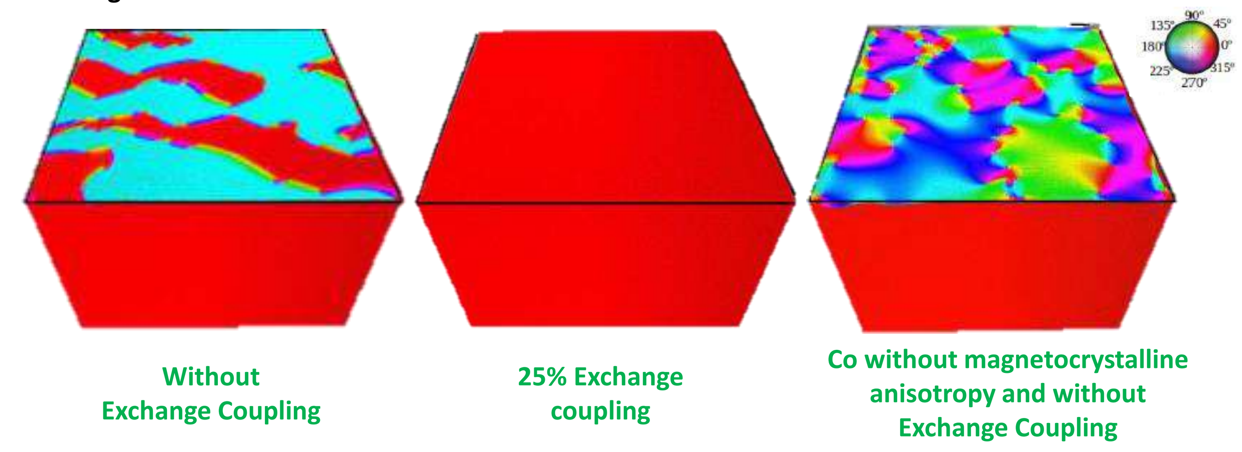


However, it is observed that the easy axis of the cobalt layer is the same as the in-plane easy axis of the hexaferrite layer, with domains pointing either parallel or antiparallel to the underlying hard domains.

The most likely explanation is that both directions (but not sense) of the magnetization in the <u>two layers are coupled structurally.</u>

MICROMAGNETIC SIMULATIONS

Micromagnetic Simulations support that the dipolar coupling is unable to impose the magnetization domain pattern of the hard layer onto the soft layer in the thickness range investigated



Without Exchange Coupling | 25% Exchange coupling | Co without magnetocrystalline anisotropy and without Exchange Coupling

CONCLUSIONS

- In order to grow strontium hexaferrite having an in-plane magnetization, the growth must be carried out in magnetron sputtering at high power (260W).
- Mössbauer spectroscopy allowed the determination of the magnetization orientation of the films.
- The XRD data have shown that the films are textured and that their structural orientation changes with the sputtering power and thickness.
- Both techniques data suggest that the magnetization of the SFO films is oriented along the c-axis direction, due to the magnetocrystaline anisotropy.
- The orientation of the samples has been confirmed by XDR and Raman.
- Vector magnetization maps show the same uniaxial easy axis for both the SFO film and Co layer. However, the magnetic domains in the cobalt overlayer are not correlated with the magnetic domains in SFO surface. There exists a structural coupling between them.
- This is further supported by comparison with micromagnetic simulations, which confirm that dipolar interactions alone do not lead to an alignment of the soft spins with the hard layer magnetization.

REFERENCES

[1] G. D. Soria et al, J. Phys. D: Appl. Phys. **53** 344002 (2019)
[2] G. D. Soria et al, J. Phys. D: Appl. Phys. **54** 054003 (2020)

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