Experimental report

07/09/2022

Proposal: **5-54-372** **Council:** 4/2021

Title: Magnetic proximity effects and spin charge conversion enhancements inferromagnetic/antiferromagnetic/normal metal heterostructures

Research area: Physics

This proposal is a new proposal

Main proposer: **Peter MILLS**

Experimental team: Peter MILLS
Niklas LANGER

Local contacts: Thomas SAERBECK

Samples: Pt/NiO/CoFe2O4
Pt/NiO/Co2FeSi

Instrument	Requested days	Allocated days	From	To
D17	5	3	21/09/2021	24/09/2021

Abstract:

Normal Metal (NM)/ferromagnet(FM) heterostructures exhibit unique magnetic and electronic properties of high interest for spintronics and device applications. These systems host both the spin-Hall effect and inverse spin Hall effect and have led to the observation of a new type of magnetoresistance knows as spin Hall magnetoresistance(SMR). Recently it has been observed that the introduction of an antiferromagnetic (AFM) layer into this system can dramatically increase the spin charge conversion. One of the main assumptions in the NM/AFM/FM systems is that magnetism is spatially confined to the FM and AFM. This proposal aims to continue to work of proposal 5-54-338 by revealing the effect of the AFM layer on magnetic proximity effects in the normal metal, which is critical for pure spin conversion. Polarized neutron reflectometry provides the required high interface and magnetic sensitivity to identify and quantify the magnetization profile of the FM and nonmagnetic metal and AFM layers.

Experiment report for proposal 5-54-372: **Magnetic proximity effects and spin charge conversion enhancements in ferromagnetic/antiferromagnetic/normal metal heterostructures**

Introduction

Normal Metal (NM)/ferromagnet(FM) heterostructures exhibit unique magnetic and electronic properties of high interest for spintronics and device applications. These systems host both the spin-Hall effect and inverse spin Hall effect and have led to the observation of a new type of magnetoresistance known as spin Hall magnetoresistance(SMR)[1]. Recently it has been observed that the introduction of an antiferromagnetic (AFM) layer into this system can dramatically increase the spin charge conversion[2]. One of the main assumptions in the NM/AFM/FM systems is that magnetism is spatially confined to the FM and AFM. This proposal aimed to continue to work of proposal 5-54-338 by revealing the effect of the AFM layer on magnetic proximity effects in the normal metal, which is critical for pure spin conversion. Polarized neutron reflectometry (PNR) provides the required high interface and magnetic sensitivity to identify and quantify the magnetization profile of the FM and nonmagnetic metal and AFM layers.

Methodology

We aimed to investigate if magnetic proximity effects (MPEs) were present in the Pt layer of heterostructure of FM/AFM/NM and FM/NM and to correlate this with SMR and spin Seebeck effect (SEE) measurements. To do this thin film heterostructures IEA18 CFS/Pt, IEA15 CFS/NiO/Pt, and AK44 CFS/NiO/Pt, have been grown on MgO substrates at the University of York via molecular beam epitaxy (MBE) and pulsed laser deposition (PLD).

Samples have been measured with both PNR and XRR. The PNR experiments were performed at the D17 instrument (ILL, Grenoble, France) in polarized time-of-flight (TOF) mode. Sample temperature and magnetic fields were controlled by an Oxford Instruments 7 T vertical field cryomagnet. Neutrons with wavelengths 4-20 Å were used to ensure the constant polarization of $p_0 > 99\%$.

NiO ordering temperature T_N is typically 535 K, however due to finite size effects, this value may be lower for our heterostructures[1]. PNR measurements were taken at RT and 5 K to observe any changes in NiO magnetization. Fields of B=0.1 T were used as they are sufficiently strong enough to saturate the CFS layer, but remain low enough such that any detected magnetization in the Pt layer can be attributed to MPEs and not a paramagnetic response from Pt.

Results

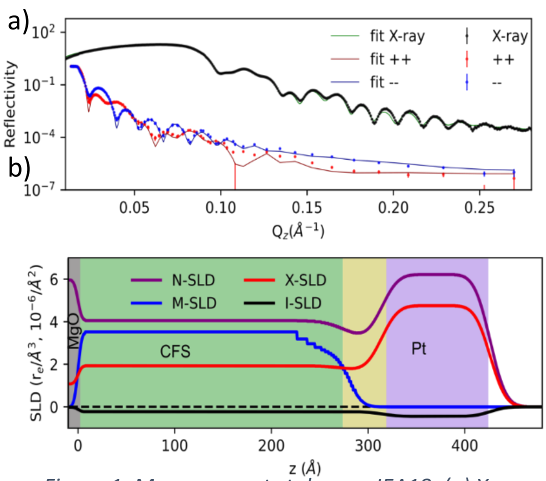

Figure 1: Measurements taken on IEA18. (a) X-ray and polarised neutron reflectivity vs momentum transfer vector normal to the sample surface (Qz). (b) Neutron scattering length density (N-SLD), Magnetic-SLD (MSLD), X-ray SLD (X-SLD) and imaginary SLD (I-SLD) vs sample depth, with z=0 at

PNR measurements of IEA18 at 0.1 T and 5 K are shown in figure 1. The simplest model was a layer of CFS, an intermediate layer, and then a layer of Pt.

Pt thickness was found to be 113 Å ± 1 Å. The Intermediate region (yellow) presented a thickness of 33 Å ± 7 Å. No magnetization was present in this layer. This layer showed a lower scattering length density (SLD) than both CFS and platinum for both X-rays and neutrons, with the relative reduction being the largest for neutrons.

In CFS three magnetic regions were simulated, starting from the substrate they are 120 Å, 121 Å and 40 Å in length. The first two magnetic regions present magnetization of 6.1 μ_B /F.U. ± 0.1 μ_B /F.U. and 6.1 μ_B /F.U. ± 0.8 μ_B /F.U.. In the final 40 Å region, the magnetization is 4.15 μ_B /F.U. ± 0.4 μ_B /F.U..

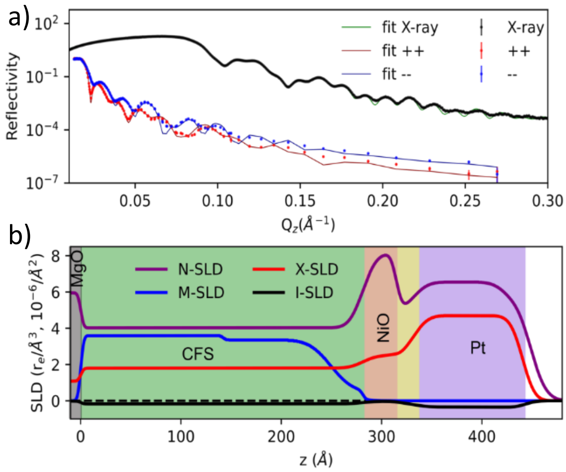

Figure 2: Measurements taken on IEA15. a) X-ray and polarised neutron reflectivity vs momentum transfer vector normal to the sample surface (Qz) . b) N-SLD, MSLD, X-SLD and I-SLD vs sample depth, with z=0 at the surface of the substrate.

PNR measurements of IEA15 at 0.1 T and 5 K are shown in figure 2. The simplest model was a layer of CFS, followed by a layer of NiO, then an intermediate layer, a layer of Pt and a surface layer. The Pt layer was found to be 104 Å ± 3 Å. The CFS/NiO chemical interface RMS roughness is 11 Å ± 1 Å .

The intermediate layer thickness is 16 Å ± 6 Å. For the neutron SLD, this layer shows a reduced SLD with respect to (w.r.t) both NiO and Pt. For the X-ray SLD, this layer is less visible. This is due to the lower SLD of NiO w.r.t Pt. The effect of the lower X-SLD of the intermediate layer is to increase the steepness of the roughness profile at the NiO/Pt interface. This does not affect the fit or model greatly, meaning x-rays have a low sensitivity to this layer.

Three independent magnetic layers were simulated in CFS. Starting from the substrate they are 145 Å, 108 Å, and 32 Å in length. CFS magnetization in the first region is 6.23 μ_B /F.U. ± 0.3 μ_B /F.U., second is 5.8 μ_B /F.U. ± 0.2 μ_B /F.U. and in the last region is 0.2 μ_B /F.U. ± 0.2 μ_B /F.U. . In the final 32 Å region there is a clear decreasing magnetization gradient leading up to the CFS/NiO interface.

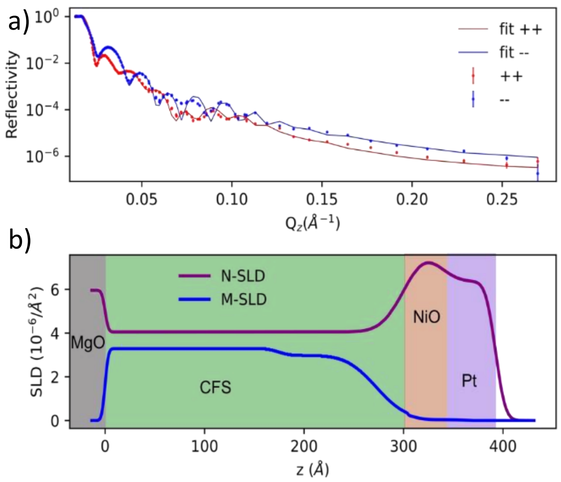

Figure 3: Measurements taken on AK44. a) Shows polarised neutron reflectivity vs momentum transfer vector normal to the sample surface (Qz) . b) Scattering length density (SLD) and Magnetic-SLD (MSLD) vs sample depth, with z=0 at the surface of the subs

PNR measurements of AK44 at B = 0.1 T and T = 5 K, are shown in figure 3. The simplest model was a layer of CFS, followed by a layer of NiO and then a layer of Pt. The Pt layer was found to be 61 Å ± 6 Å. The CFS/NiO chemical interface RMS roughness is 20.0 Å ± 2 Å and the NiO/Pt RMS roughness is 16 Å ± 4 Å.

Three independent magnetic layers were simulated in CFS. Starting from the substrate they are, 175 Å, 98 Å and 32 Å. CFS magnetization in the first region is 5.7μ_B /F.U. ± 0.1μ_B /F.U., second is 5.2 μ_B /F.U. ± 0.1 μ_B /F.U. and in the last region is 0.1 μ_B /F.U. ± 0.4μ_B /F.U..

Discussion

All CFS films show a steep magnetic gradient of around 50 Å top interface. This gradient is likely due to oxidation of the CFS layer, with the oxygen diffusing from the NiO in IEA15 and AK44, and from surface contamination in IEA18 from time outside of ultra-high vacuum before Pt deposition. This reduced magnetization at either the CFS/NiO or CFS/Pt interface will have significantly reduced any MPEs.

IEA18 and IEA15 showed similar spin voltages (results not shown) of 7 ± 1 nV.m/W and 5 ± 1 nV.m/W, respectively, and AK44 shows the highest spin voltage of 12±1 nV.m/W.

The bulk magnetization of CFS is 6 μ_B /F.U. [3]. The first magnetic regions of IEA18 and IEA15 present magnetizations of 6.1 μ_B /F.U. ± 0.1 μ_B /F.U. and 6.23 μ_B /F.U. ± 0.3 μ_B /F.U., respectively, in agreement with CFSs bulk magnetization. The magnetization in AK44 in the first magnetic region is 5.7 μ_B /F.U. ± 0.1 μ_B /F.U., slightly lower than the bulk value of CFS. Given that AK44 has the largest spin voltage, it is unlikely the small decrease in CFS magnetization has a large effect on the measured SEE. AK44 does not present an intermediate layer of decreased SLD at the lower interface of Pt, unlike IEA15 and IEA18. The Pt layer on AK44 was found to be 61 Å ± 6 Å, thinner than both IEA15 with 104 Å ± 3 Å and IEA18 with 113 Å ±1 Å. It is expected that the reduced Pt layer thickness in AK44 lead to the comparatively increased spin voltage, but it's also possible that the improved lower Pt interface had an effect.

Future outlook

To improve this experiment the reduced magnetization at the surface of CFS must be increased so that MPEs can potentially be detected. One option would be to improve the growth procedure of the CFS/NiO/Pt structures. This could be achieved by deposing a thin metallic layer of, for example Fe or Ni, on the surface of CFS before NiO deposition. This layer will help to reduce the amount of oxygen that defuses into the CFS layer during NiO deposition. To avoid any layers of surface contamination on the NiO or CFS surface, the depositions should all be performed in situ.

Another option is to remove the risk of oxidization entirely by using a magnetic ferrite in place of CFS. Ferrites are typically at their ambient and so will not oxidize. Interesting candidates could be either $CoFe_2O_4$, a hard magnet and insulating, or Fe3O4, a potential half-metal with a large theoretical magnetization of 4 μ_B /F.U.. The new sample structures would be either CoFe2O4/NiO/Pt or Fe3O4/NiO/Pt.

References

[1] H. Nakayama, et al., PRL. 110, 206601 (2013)

[2] W. Lin et al. PRL 118, 067202 (2017)

[3] S. Wurmehl, et al. APL. 88, 032503 (2006)