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Heusler Alloys with bcc Tungsten Seed Layers for GMR Junctions

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Abstract

We demonstrate that polycrystalline Co$_2$FeSi Heusler alloys films can be grown with perpendicular anisotropy without the use of an MgO interface. By heating the substrate to 400 °C prior to deposition and using a tungsten seed layer perpendicular anisotropy is induced in the Heusler layer. This is maintained as the thickness of the Co$_2$FeSi is increased up to 12.5 nm. The layers with thickness dependent coercivity can be implemented into a giant magnetoresistance structure leading to spin-valve behaviour without the need for an exchange biased pinned layer.

Keywords

- Heusler Alloys
- Perpendicular Anisotropy
- Spintronics
- Spin-valves

1. Introduction

The advancement of spintronic devices requires the further minimisation of device dimensions and signal outputs while reducing device power consumption and heating via reduction of the resistance area product RA[1–4]. This is especially true in the instance of magnetic tunnel junctions (MTJs) where the tunnelling barrier is an insulator and therefore R is unavoidably high. The solution is to transfer back to giant magnetoresistive (GMR) devices with low R but these devices have a far lower signal-to-noise ratio (SNR) due to a smaller GMR ratio compared to a tunnelling magnetoresistance (TMR) ratio [1, 2, 5]. Another obstacle is the implementation of perpendicular magnetic anisotropy (PMA) in junctions which has been realised by the insertion of the MgO layer, where hybridisation of the O-p orbitals with neighbouring layers generates the PMA in MTJs [6, 7].

Heusler alloys have attracted attention in spintronics due to high Curie temperatures and predicted 100% spin polarisations [2, 8–13]. Since TMR and GMR are spin dependent phenomena the Heusler alloys allow for potentially infinite MR ratios and are hence exceedingly attractive. These alloys are, however, cubic and as such possess little magnetocrystalline anisotropy [12]. Therefore thin films have anisotropy in-plane which is non-ideal for the

latest applications. This could be overcome as in conventional MTJs by using an MgO/Co-based Heusler interface but this is contradictory to the low RA target [7]. Instead crystallographic manipulation can be implemented via seed layers. If sufficient strain is induced in the Heusler alloy then PMA can be achieved without an MgO layer. Additionally interfaces with heavy metals have been found to induce strong anisotropy in ferromegnetic layers, however the origins of this are largely unknown [14, 15]. Therefore GMR devices with Heusler alloys with PMA can be a strong competitor for MTJs.

Previous work has shown that a vanadium seed layer can induce PMA in Co$_2$FeSi. Furthermore a second vanadium interface can maximise this effect leading to a film with a weak but dominant PMA [16]. In this work we have used a tungsten seed layer for Co$_2$FeSi layers of increasing thickness. Deposition on heated substrates has been used to crystallise the films. The films have been characterised crystallographiically and magnetically to investigate the magnetic anisotropy. The optimised condition has then been implemented into GMR multilayers and further characterised. Co$_2$FeSi was used as the ferromagnetic Heusler alloy due to a saturation magnetisation M_S of 1200 emu/cm^3 and a Curie temperature T_C of 1100 K and to easily compare to the anisotropy induced with a V seed layer [12, 16, 17].

2. Materials and Methods

Samples were deposited on Si(001) substrates using a PlasmaQuest High Target Utilisation Sputtering system (HiTUS) with a bias voltage of -900 V and a process pressure of 1.86 mTorr in order to maximise grain volume [18]. Prior to deposition the native SiO$_2$ layer was removed by exposure to the plasma. Substrates were heated to a temperature of 400 °C in order to encourage crystallisation of the Heusler alloy [7, 19–21]. Tungsten was chosen as a seed layer for the Heusler alloy due to

a large lattice mismatch of 22.2% and a high spin-orbit coupling. An initial sample consisiting of Si(001)//W (20 nm) was deposited to investigate the crystallisation of the seed layer. Subsequently Co_2FeSi was deposited from a stoichiometric target in varying thicknesses for samples with the structure Si(001)//W (10 nm)/Co_2FeSi (2.5 nm to 12.5 nm)/Ru (3 nm). Finally these Co_2FeSi layers were incorporated into a spin-valve structure with a thin W barrier Si(001)//W (10 nm)/Co_2FeSi (12.5 nm)/W (0.4 nm to 0.6 nm)/Co_2FeSi (12.5 nm)/Ru (3 nm).

The samples were magnetically characterised using ADE Model 10 and Lakeshore 7300 vibrating sample magnetometers (VSM) with a sensitivity of 1×10^{-6} emu and a field precision of 1 Oe. The sample mounts are rotational allowing for measurement both in-plane and out-of-plane. For crystallographic analysis a Rigaku SmartLab X-ray diffractometer (XRD) was used with a Cu- source. A 6-axis goniometer was used to measure samples in both in-plane and out-of-plane geometries.

3. Results and Discussion

Figure 1a shows the $\vartheta/2\vartheta$ and $2\vartheta_\chi$ scans for the 20 nm layer of W. Several crystal planes of W are visible including the {100}, {110} and {211} planes, with the {110} peak having a dominant magnitude. This implies a significant crystallisation in the W layer but a lack of 2D texture. Scherrer analysis of the {110} reflection gives a crystallite size on the order of 8 nm. However, the fitted X-ray Reflectivity (XRR) data shown in fig. 1b gives a 94% dense film with a roughness of 0.2 nm.
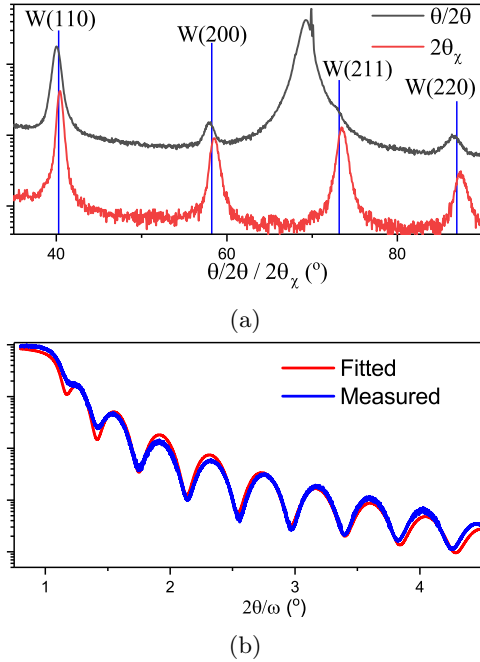

(a)

(b)

Figure 1: (a) ϑ-$2\vartheta$and $2\vartheta_\chi$ scans of the 20 nm W film and (b) the associated XRR scan.

Layers of Co_2FeSi were then added in increasing thickness t_{CFS} from 2.5 nm to 12.5 nm to a 10 nm seed layer of W. Figure 2a shown the in-plane $2\vartheta_\chi$ scan for the sample with only 2.5 nm of Co_2FeSi. Even at this low thickness the Co_2FeSi has crystallised in the {110} direction. The multiple reflections from the W layer are still present so there is no dominant 2D texture in the film. The in-plane Co_2FeSi {110} plane spacing is reduced to 3.98 Å which is 2% smaller than the out-of-plane value. A strain has been induced at the W/Co_2FeSi interface resulting in a slight tetragonal distortion, with a long axis out-of-plane. This is a possible origin of the anisotropy induced in the films. Scherrer analysis of the Co_2FeSi {110} reflections indicates a small increase in crystallite size from 7.8 nm to 9.7 nm in the 2.5 nm and 12.5 nm films respectively.
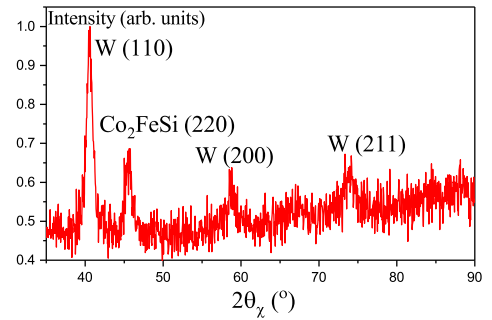
(a) $2\theta_\chi$ scan of a 2.5 nm layer of Co_2FeSi on a 10 nm seed layer of W.
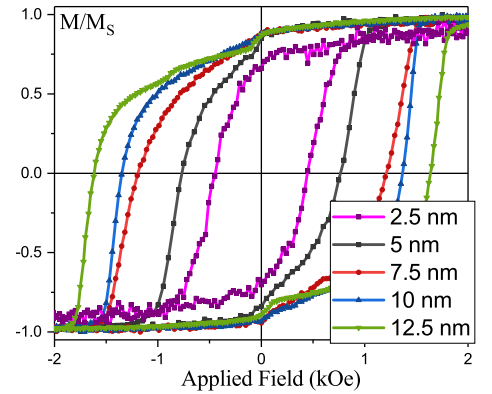
(b) Out-of-plane M-H loops for increasing Co_2FeSi thicknesses.

Figure 2: (a) Crysatllographic and (b) magnetic characterisation of increasing Co_2FeSi layer thicknesses.

The magnetic measurements out-of-plane are shown in fig. 2b. All of the films exhibit a perpendicular magnetic anisotropy regardless of t_{CFS}. The saturation magnetisation increases with t_{CFS} from 480 emu/cm^3 to 605 emu/cm^3 as shown in fig. 3a. This is potentially due to magnetically dead layers at the interfaces, which become less significant as t_{CFS} increases. Figure 3a also shows that the coercivity is also seen to increase with the film thickness up to a maximum of 1.65 kOe in the 12.5 nm film.

The reversal mechanism of the films has the domain

nucleation characteristics expected of a film with high intergranular exchange coupling. However this only occurs after significant domain rotation which reduces the magnetisation to half of M_S. After this point the reversal is controlled by domain wall motion and pinning. The exact value at which the reversal mechanism changes is not possible to determine. However the level of rotation seems to increase with t_{CFS} before nucleation is achieved. There are two causes for the increase in magnetisation rotation. Firsty this indicates an increasing magnetic anisotropy where the anisotropy energy overcomes the intergranular exchange coupling which favours nucleation reversal. As such this suggests that the perpendicular magnetic anisotropy is increasing with t_{CFS}. Secondly both coercivity and grain size increase with t_{CFS}. The larger grains in the film allow for a greater degree of rotation in the film before reversal. However the increase in the grain size is only small and has a small impact on the coercivity.

A direct measure of the anisotropy using a coherent reversal mechanism is not possible due to the intergranular coupling. The coupling leads to a low but non-zero coercivity hysteresis loop in the plane of the sample. As such the anisotropy field cannot be determined and thus the anisotropy cannot be calculated from the M-H data. Further investigation using a technique such as ferromagnetic resonance (FMR) must be carried out to obtain a numerical value of K.

Figure 3b shows the out-of-plane DC demagnetising curve for the sample $t_{CFS} = 12.5$nm and the calculated irreversible susceptibility χ_{irrev}. Due to the intergranular coupling field the demagnetising field H_D cannot be accounted for, as a correction factor of 4π results in an overcorrection. The DCD curve therefore can only be measured at the effective zero field inclusive of H_D. However, for device application this is the real field that the sample will encounter and as such is a valid method to determine an effective activation volume V_{eff}. From time dependence measurements $S(H)$ was determined and hence V_{eff}. For the 12.5 nm film the activation volume diameter D_{eff} was found to be (15 ± 3) nm. This is almost double the crystallite size in the film and supports the presence of strong intergranular coupling in the film.

Since the coercivity of the layers is strongly dependent on t_{CFS} a spin valve structure can be constructed where the layers switch at very different fields, ideal for creating a GMR structure. Figure 4 shows the M-H data out-of-plane for spin-valve structures based on the structure Si(001)//W (10 nm)/Co$_2$FeSi (12.5 nm)/W (0.4 nm to 0.6 nm)/Co$_2$FeSi (12.5 nm)/Ru (3 nm). The two Co$_2$FeSi layers of the structure switch distinctly with the W 0.5 nm and 0.6 nm layers which is ideal for a spin-valve structure. The coercivity of the films is also greater than that seen in the individual layers. This implies some coupling between the two ferromagnetic layers through the W layer. The W layer thickness was set at the peak of the antiferromagnetic coupling strength [22], however there is no clear evidence that this exact coupling is observed.
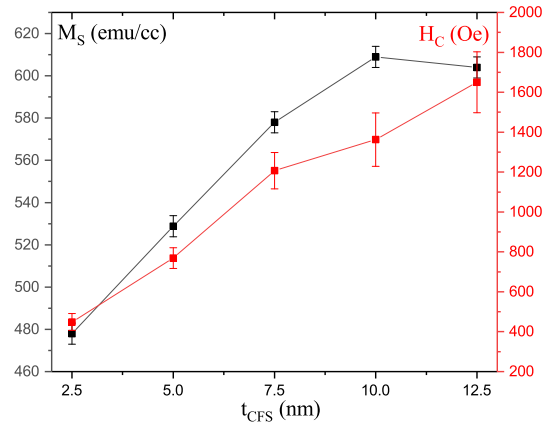
(a) The increase of M_S and H_C with the thickness of the magnetic layer t_{CFS}.
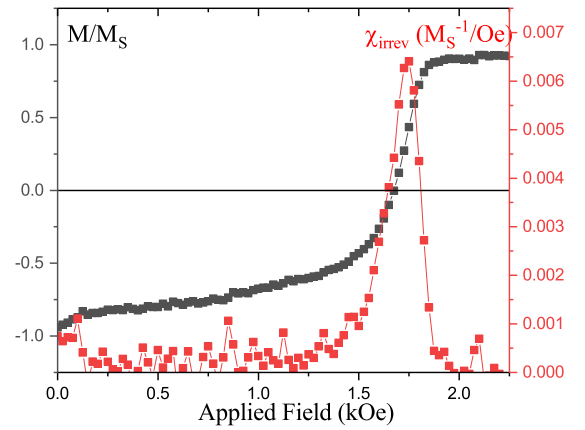
(b) DCD curve of a 12.5nm Co$_2$FeSi film out-of-plane and the calculated χ_{irrev}.
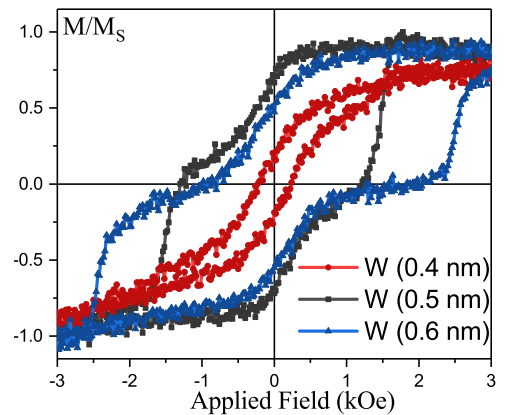

Figure 4: Out-of-plane M-H loops for spin-valve structures based on Co$_2$FeSi with varying thickness of W barrier layer.

4. Conclusions

We have shown that interfacial perpendicular magnetic anisotropy (PMA) in Co_2FeSi Heusler alloys can be induced by addition of a tungsten seed layer. PMA can be maintained and increases up to a Co_2FeSi layer thickness of 12.5 nm, a thickness much greater than usually observed [7, 16]. Spin-valve structures were also deposited using the optimised $W/Co_2FeSi/W/Co_2FeSi$ structure to maximise PMA, where switching clearly depends upon the Co_2FeSi layer thickness as is desirable for applications.

5. Acknowledgements

[1] K. Nagasaka, CPP-GMR technology for magnetic read heads of future high-density recording systems, Journal of Magnetism and Magnetic Materials 321 (6) (2009) 508–511. doi:10.1016/j.jmmm.2008.05.040.

[2] Z. Diao, M. Chapline, Y. Zheng, C. Kaiser, A. Ghosh Roy, C. J. Chien, C. Shang, Y. Ding, C. Yang, D. Mauri, Q. Leng, M. Pakala, M. Oogane, Y. Ando, Half-metal CPP GMR sensor for magnetic recording, Journal of Magnetism and Magnetic Materials 356 (2014) 73–81. doi:10.1016/j.jmmm.2013.12.050. URL http://dx.doi.org/10.1016/j.jmmm.2013.12.050

[3] S. Mitani, Spin-transfer magnetization switching in ordered alloy-based nanopillar devices, Journal of Physics D-Applied Physics 44 (38). doi:10.1088/0022-3727/44/38/384003.

[4] T. M. Nakatani, A. Rajanikanth, Z. Gercsi, Y. K. Takahashi, K. Inomata, K. Hono, Structure, magnetic property, and spin polarization of Co2FeAlxSi1x Heusler alloys, Journal of Applied Physics 102 (3) (2007) 033916–033916–8. doi:doi:10.1063/1.2767229. URL http://jap.aip.org.libproxy1.nus.edu.sg/resource/1/japiau/v102/i3/p033916{_}s1{%}5Cnhttp://scitation.aip.org.libproxy1.nus.edu.sg/getpdf/servlet/GetPDFServlet?filetype=pdf{&}id=JAPIAU000102000003033916000001{&}idtype=cvips{&}doi=10.1063/1.2767229{&}prog=normal

[5] S. M. Thompson, The discovery, development and future of GMR: The Nobel Prize 2007, Journal of Physics D: Applied Physics 41 (9) (2008) 93001. doi:10.1088/0022-3727/41/9/093001. URL http://stacks.iop.org/0022-3727/41/i=9/a=093001?key=crossref.e81e7c8f22afb14e30b406657fab7e50

[6] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, H. Ohno, A perpendicular-anisotropy CoFeBMgO magnetic tunnel junction, Nature Materials 9 (9) (2010) 721–724. doi:10.1038/nmat2804. URL http://www.nature.com/doifinder/10.1038/nmat2804

[7] Y. Cui, B. Khodadadi, S. Sch??fer, T. Mewes, J. Lu, S. A. Wolf, Interfacial perpendicular magnetic anisotropy and damping parameter in ultra thin Co2FeAl films, Applied Physics Letters 102 (16). doi:10.1063/1.4802952. URL https://doi.org/10.1063/1.4802952http://aip.scitation.org/toc/apl/102/16

[8] T. M. Nakatani, N. Hase, H. S. Goripati, Y. K. Takahashi, T. Furubayashi, K. Hono, Co-Based Heusler Alloys for CPP-GMR Spin-Valves With Large Magnetoresistive Outputs, IEEE Transactions on Magnetics Transactions on Magnetics 48 (5) (2012) 1751–1757. doi:10.1109/tmag.2011.2174436.

[9] K. Nikolaev, P. Kolbo, T. Pokhil, X. Peng, Y. Chen, T. Ambrose, O. Mryasov, " All-Heusler alloy " current-perpendicular-to-plane giant magnetoresistance, Applied Physics Letters 94 (101). doi:10.1063/1.3126962. URL http://dx.doi.org/10.1063/1.3126962http://aip.scitation.org/toc/apl/94/22http://dx.doi.org/10.1063/1.3126962{%}0Ahttp://aip.scitation.org/toc/apl/94/22

[10] T. Seki, Y. Sakuraba, H. Arai, M. Ueda, R. Okura, H. Imamura, K. Takanashi, High power all-metal spin torque oscillator using full Heusler Co2(Fe,Mn)Si, Applied Physics Letters 105 (9). doi:10.1063/1.4895024. URL http://dx.doi.org/10.1063/1.4895024http://aip.scitation.org/toc/apl/105/9

[11] Y. Sakuraba, S. Kokado, Y. Hirayama, T. Furubayashi, H. Sukegawa, S. Li, Y. K. Takahashi, K. Hono, Quantitative analysis of anisotropic magnetoresistance in Co 2 MnZ and Co 2 FeZ epitaxial thin films: A facile way to investigate spin-polarization in half-metallic Heusler compounds, Applied Physics Letters 1041 (10) (2014) 172407–7. doi:10.1063/1.4874851. URL http://dx.doi.org/10.1063/1.4874851

[12] T. Graf, C. Felser, S. S. P. Parkin, Simple rules for the understanding of Heusler compounds, Progress in Solid State Chemistry 39 (1) (2011) 1–50. doi:10.1016/j.progsolidstchem.2011.02.001. URL http://dx.doi.org/10.1016/j.progsolidstchem.2011.02.001

[13] Y. K. Takahashi, N. Hase, M. Kodzuka, A. Itoh, T. Koganezawa, T. Furubayashi, S. Li, B. S. D. C. S. Varaprasad, T. Ohkubo, K. Hono, Structure and magnetoresistance of current-perpendicular-to-plane pseudo spin valves using Co2Mn(Ga0.25Ge0.75) Heusler alloy, Journal of Applied Physics 113 (22) (2013) 0–7. doi:10.1063/1.4809643.

[14] S. Peng, M. Wang, H. Yang, L. Zeng, J. Nan, J. Zhou, Y. Zhang, A. Hallal, M. Chshiev, K. L. Wang, Q. Zhang, W. Zhao, Origin of interfacial perpendicular magnetic anisotropy in MgO/CoFe/metallic capping layer structures, Scientific Reports 5 (1) (2015) 18173. arXiv:1506.04078, doi:10.1038/srep18173. URL http://www.nature.com/articles/srep18173

[15] Y.-W. Oh, K.-D. Lee, J.-R. Jeong, B.-G. Park, Interfacial perpendicular magnetic anisotropy in CoFeB / MgO structure with various underlayers Interfacial perpendicular magnetic anisotropy in CoFeB / MgO structure with various underlayers, Journal of Applied Physics 115 (17) (2014) 17C724. doi:10.1063/1.4864047. URL https://doi.org/10.1063/1.4864047http://aip.scitation.org/toc/jap/115/17

[16] W. Frost, A. Hirohata, Perpendicular anisotropy in heusler alloy layers induced by a V seed layer, IEEE Transactions on Magnetics 52 (7) (2016) 2–5. doi:10.1109/TMAG.2016.2517738.

[17] S. Wurmehl, G. H. Fecher, H. C. Kandpal, V. Ksenofontov, C. Felser, H.-J. Lin, J. Morais, Geometric, electronic, and magnetic structure of <math display="inline"><mrow> <msub> <mi>Co</mi> <mn>2</mn> </msub> <mi>FeSi</mi> </mrow> </math> : Curie temperature and magnetic moment measurements and calculations, Physical Review B 72 (18) (2005) 184434. arXiv:0506729, doi:10.1103/PhysRevB.72.184434. URL https://journals.aps.org/prb/pdf/10.1103/PhysRevB.72.184434https://link.aps.org/doi/10.1103/PhysRevB.72.184434

[18] M. Vopsaroiu, M. J. Thwaites, S. Rand, P. J. Grundy, K. O'Grady, Novel sputtering technology for grain-size control, IEEE Transactions on Magnetics 40 (4 II) (2004) 2443–2445. doi:10.1109/TMAG.2004.828971. URL http://ieeexplore.ieee.org/document/1325532/

[19] M. Vahidi, J. A. Gifford, S. K. Zhang, S. Krishnamurthy, Z. G. Yu, L. Yu, M. Huang, C. Youngbull, T. Y. Chen, N. Newman, Fabrication of highly spin-polarized Co 2 FeAl 0.5 Si 0.5 thin-films, Applied Physics Letters 2 (2) (2014) 46108–221912. doi:

10.1063/1.2767229.
URL https://doi.org/10.1063/1.4869798

[20] Y. Du, B. S. D. C. S. Varaprasad, Y. K. Takahashi, T. Furubayashi, K. Hono, 001 textured polycrystalline current-perpendicular-to-plane pseudo spin-valves using Co2Fe(Ga0.5Ge0.5) Heusler alloy, Applied Physics Letters 103 (20). doi:10.1063/1.4829633.
URL http://dx.doi.org/10.1063/1.4829633http://aip.scitation.org/toc/apl/103/20

[21] H. Schneider, G. Jakob, M. Kallmayer, H. J. Elmers, M. Cinchetti, B. Balke, S. Wurmehl, C. Felser, M. Aeschlimann, H. Adrian, Epitaxial film growth and magnetic properties of Co2FeSi, Physical Review B 74 (17) (2006) 174426. arXiv:0606666, doi:10.1103/PhysRevB.74.174426.
URL https://journals.aps.org/prb/pdf/10.1103/PhysRevB.74.174426http://link.aps.org/doi/10.1103/PhysRevB.74.174426

[22] S. S. P. Parkin, Systematic variation of the strength and oscillation period of indirect magnetic exchange coupling through the 3d, 4d, and 5d transition metals, Physical Review Letters 67 (25) (1991) 3598–3601. doi:10.1103/PhysRevLett.67.3598.