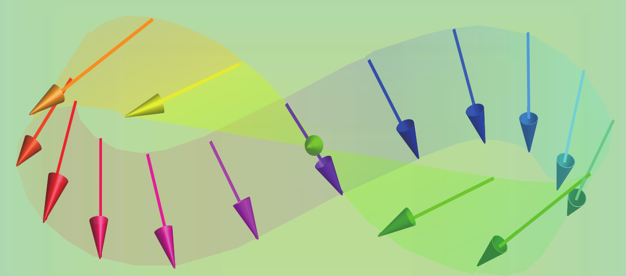
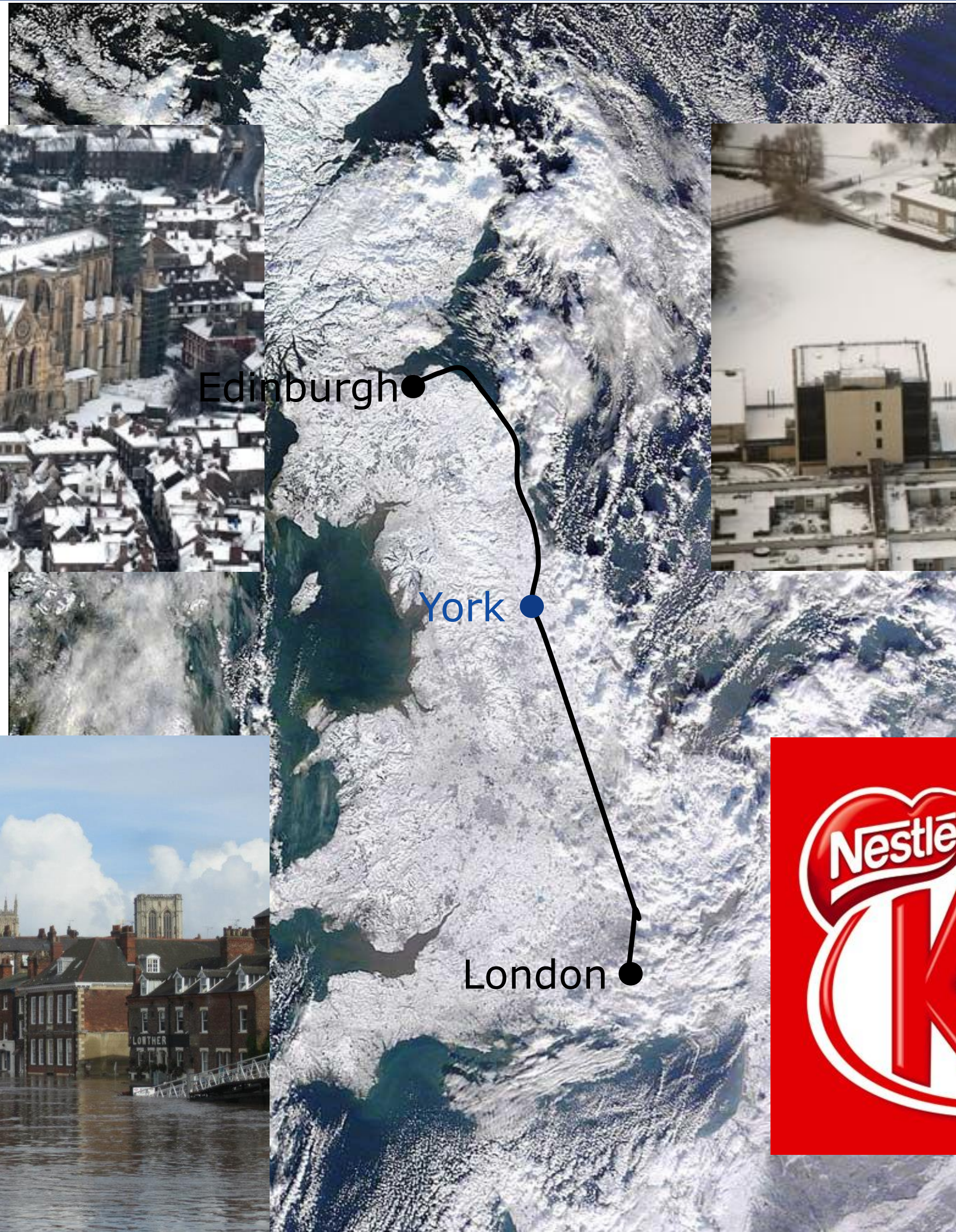
Heusler Alloy Films for Spintronic Devices



Atsufumi Hirohata

THE UNIVERSITY of York

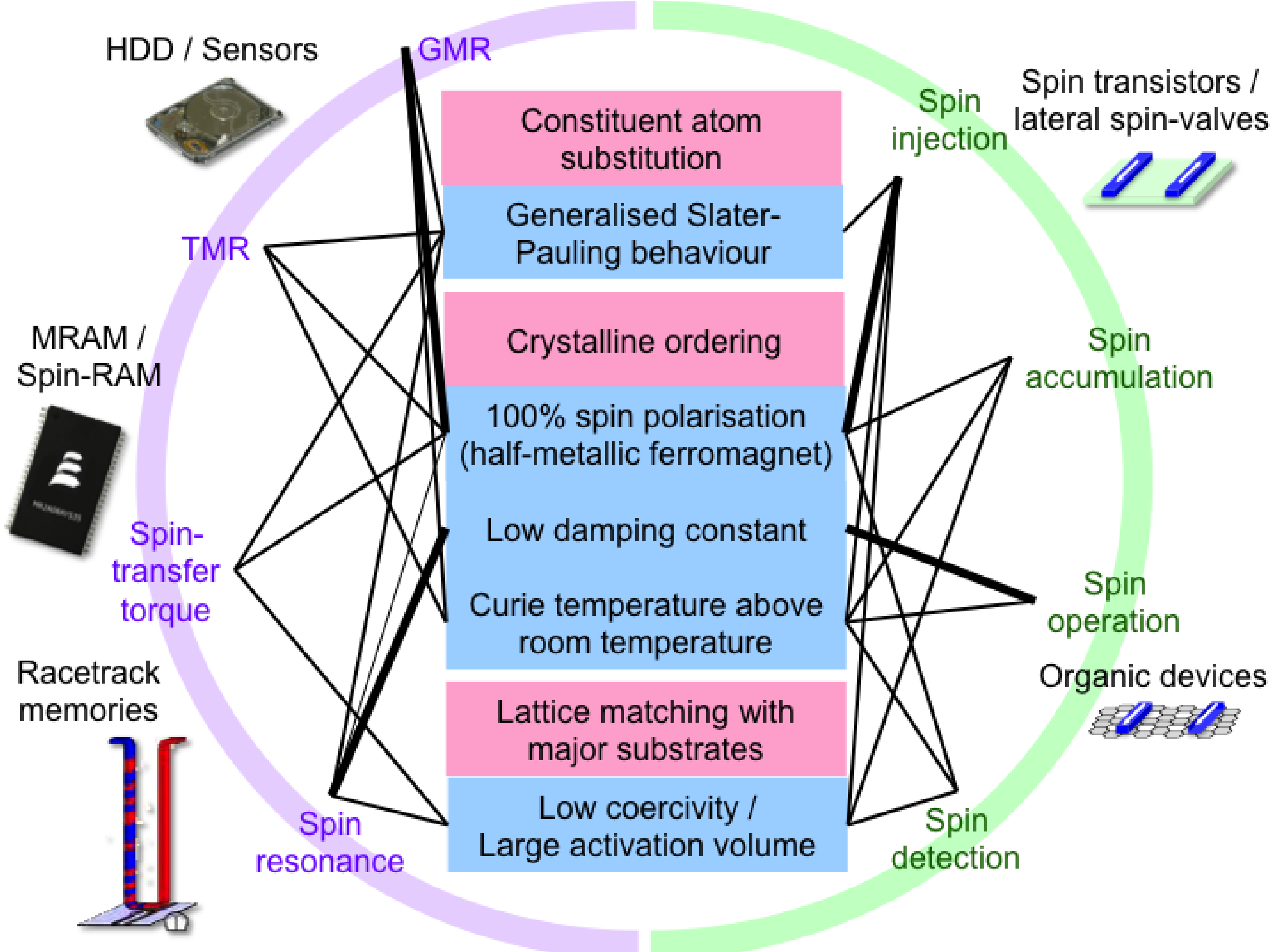
Where is York ?



* http://www.bbc.co.uk/

Applications for Heusler Alloys

Requirements for device implementations for Heusler alloys :

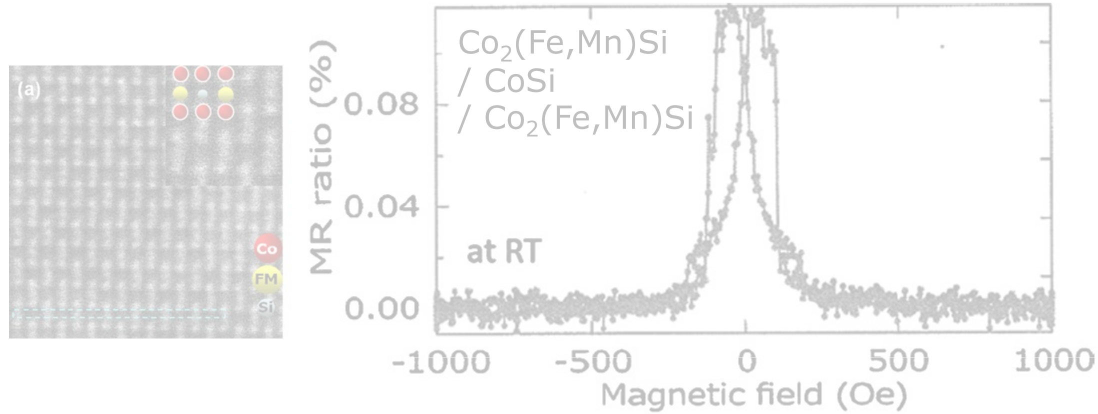


* A. Hirohata *et al.*, *Heusler Alloys* (Springer, in press).

Possible Solutions

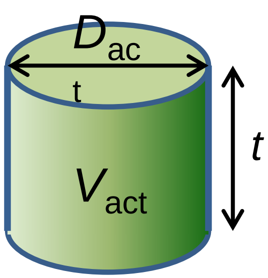
Smoothing the interfaces :

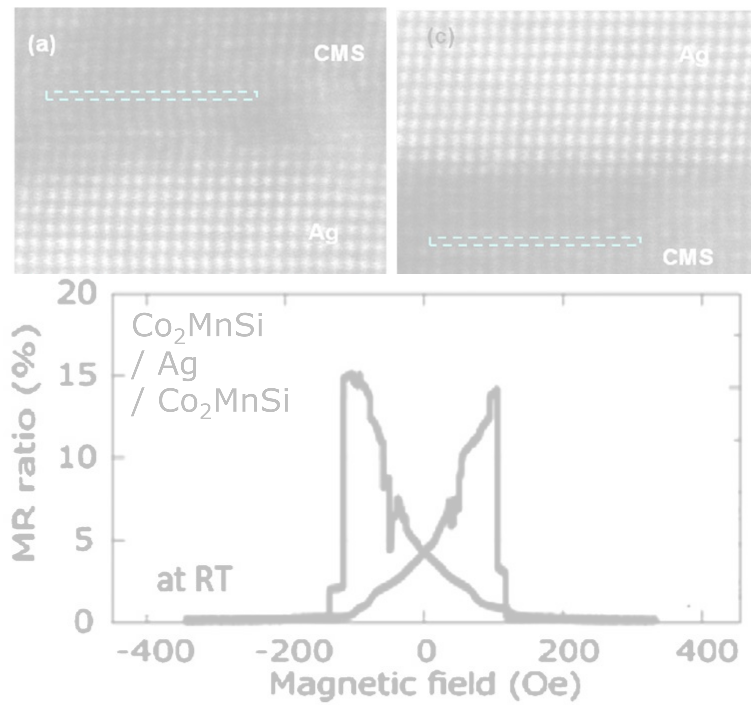
- Optimisation of the non-magnetic spacer
  → Ag
- Atomically sharp interface achieved
  → GMR ratio : ~ 15 %



Elimination of minor domains :

- Maximisation of activation volume
  → smallest volume that reverses in a single step



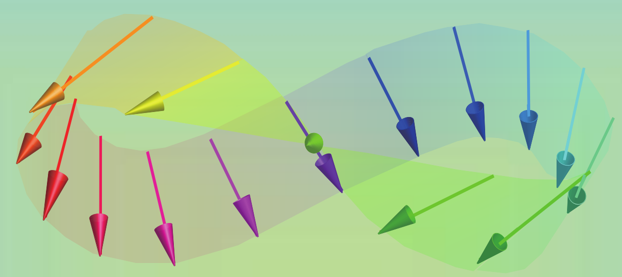


Minimisation of intermixing / deformation :

- Low-temperature annealing for Heusler alloy films
  → *in situ* TEM observation

* V. K. Lazarov *et al.*, *Appl. Phys. Lett.* 98, 242508 (2011).

Activation Volumes in Heusler Alloy Films



J. Sagar,[1] H. Sukegawa,[2] L. Lari,[1,3] V. K. Lazarov,[1] S. Mitani,[2]
K. O'Grady [1] and A. Hirohata [1]

1 The University of York 2 NIMS 3 York JEOL Nanocentre

さきがけ PRESTO

EPSRC EP/H026126/1

Engineering and Physical Sciences Research Council

Co_2FeSi Heusler-Alloy Epitaxial Film Growth

- Sample Structure:

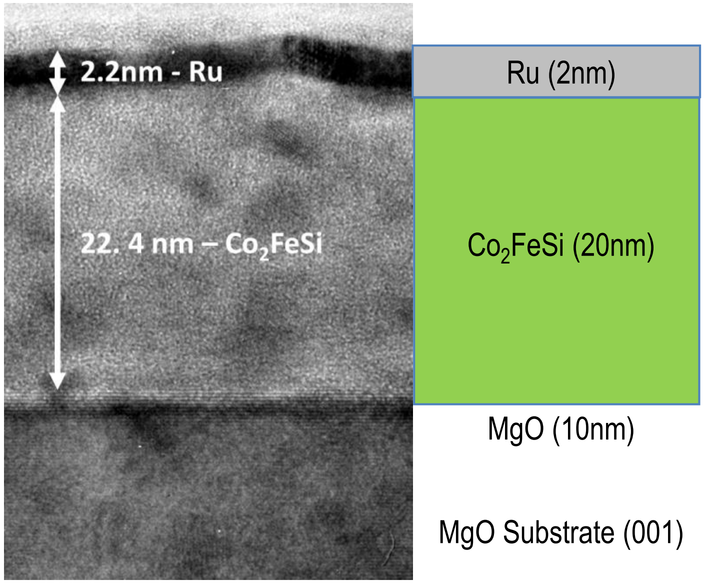
MgO [100]//MgO (10nm) / Co_2FeSi (2nm) / Ru (2nm)

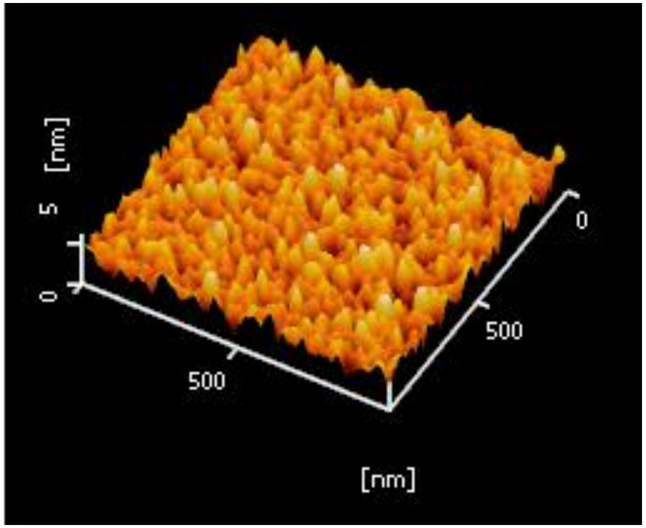
- MgO substrate annealed at 800°C before deposition of a 10 nm MgO buffer layer and annealing again at 400°C.

- Co_2FeSi deposited by UHV magnetron sputtering with base pressure of better than 3×10^{-7} Pa at a rate of 0.03 nm/s

- Post deposition AFM measurements of samples found R_a to be 4.7 nm

- *Ex-situ* post deposition annealing at 400°C, 500°C and 600°C to cause recystallisation into B2 and $L2_1$ phases.





XRD Structural Characterisation

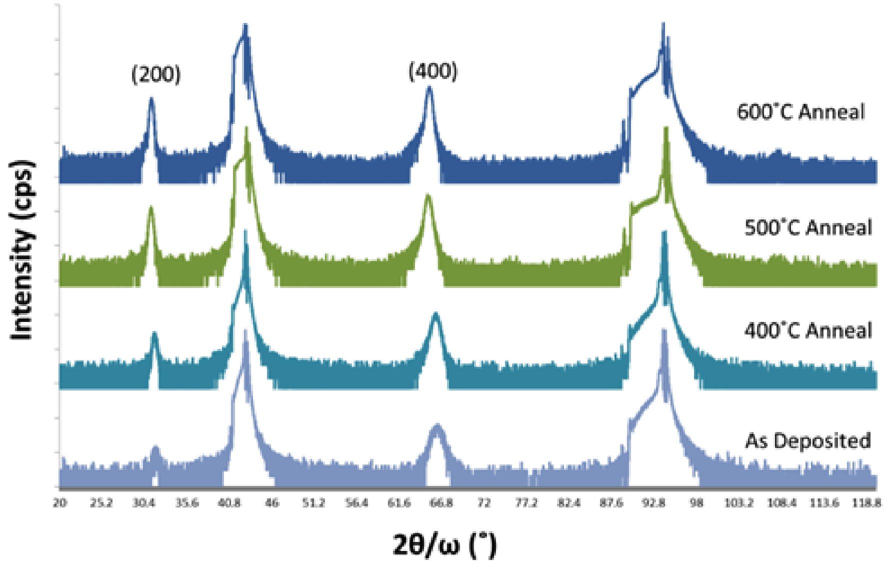
- X-ray diffraction (XRD) spectra were taken for as-deposited and post annealed films.

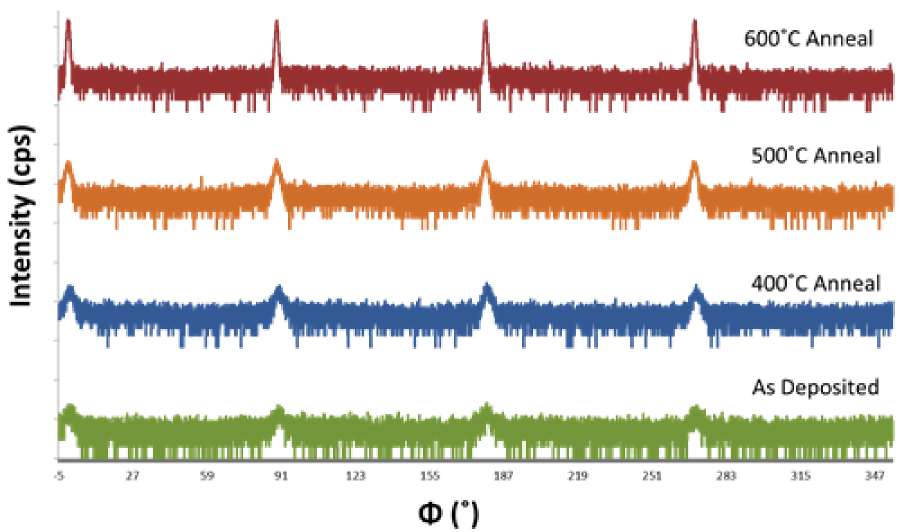
- 2θ-ω (out-of-plane) and 2θ-$\varphi\chi$ (in plane) scans have been taken.

- This allows for structural characterisation and identification of order phases.

- Increasing intensity (200) and (400) peaks are characteristic of $B2$ and $L2_1$ ordering.

- Increased intensity (111) peaks in the 2θ-$\varphi\chi$ show increased $L2_1$ ordering with increasing anneal temperature.





STEM Structural Characterisation

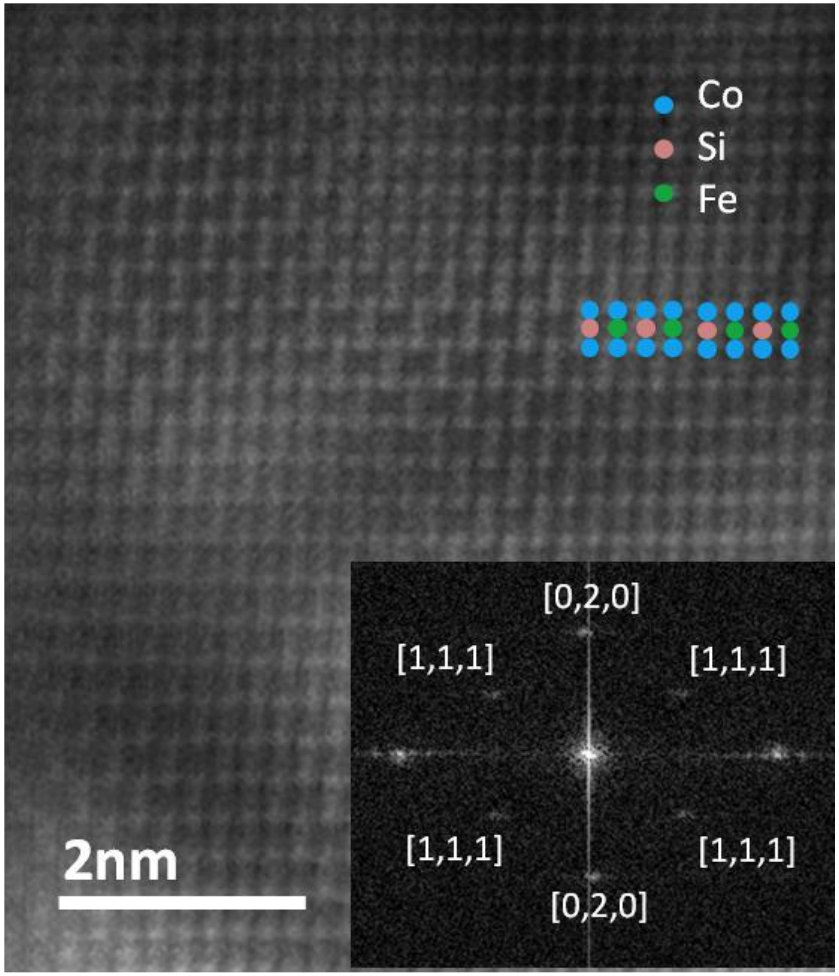
- HAADF STEM imaging with elemental contrast can be used for structural analysis of Heusler alloy films

- Using digital diffractograms and measurements of inter-atomic spacing, the in-plane and out-of-plane lattice constants have been found.

- From the [111] spots the in-plane lattice constant was found to be (5.74 ± 0.05) Å compared with the bulk value of 5.64 Å

- From the [200] spots the out-of-plane lattice constant was found to be slightly reduced at (5.44 ± 0.05) Å

- The volume of the unit cell was found to lie within error of the bulk value.



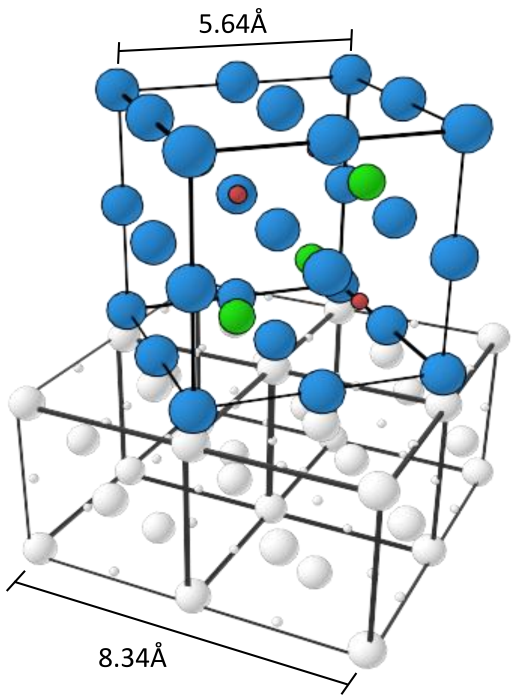
Insert reference

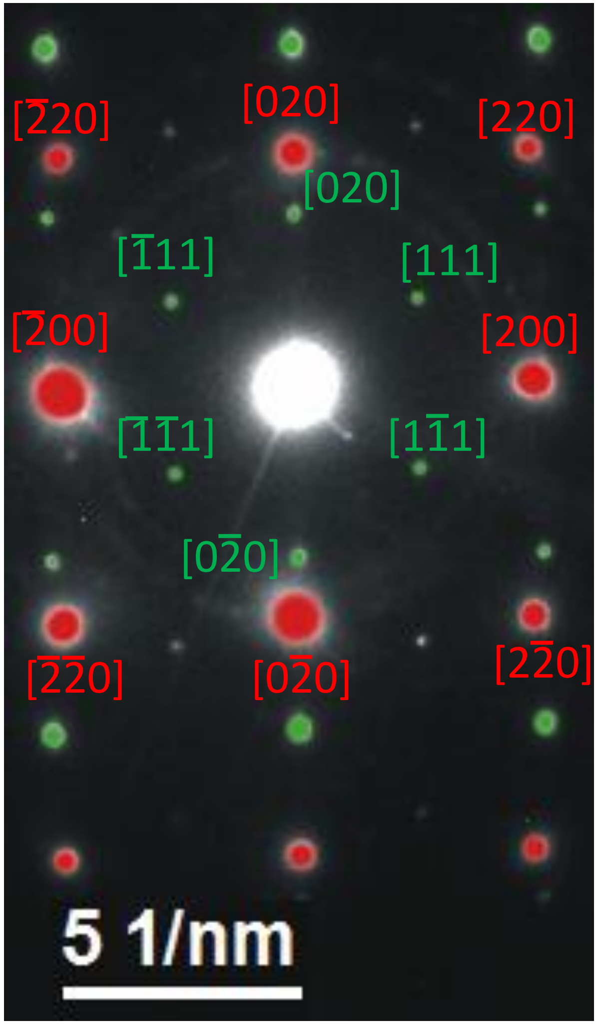
Co_2FeSi Heusler-Alloy Film Growth

- The lattice constant for Co_2FeSi is 5.64 Å and for MgO is 4.17 Å, this is a mismatch of 35%.

- To compensate for this mismatch the Co_2FeSi unit cell is found to rotate by 45° to align the Co_2FeSi [110] planes with the MgO [100].

- This rotation allows the Co_2FeSi to span two MgO unit cells and reduces the mismatch to 4.5 %.





False colour diffraction pattern showing Co_2FeSi [110] and MgO [100] reflections.

Magnetic Time Dependence

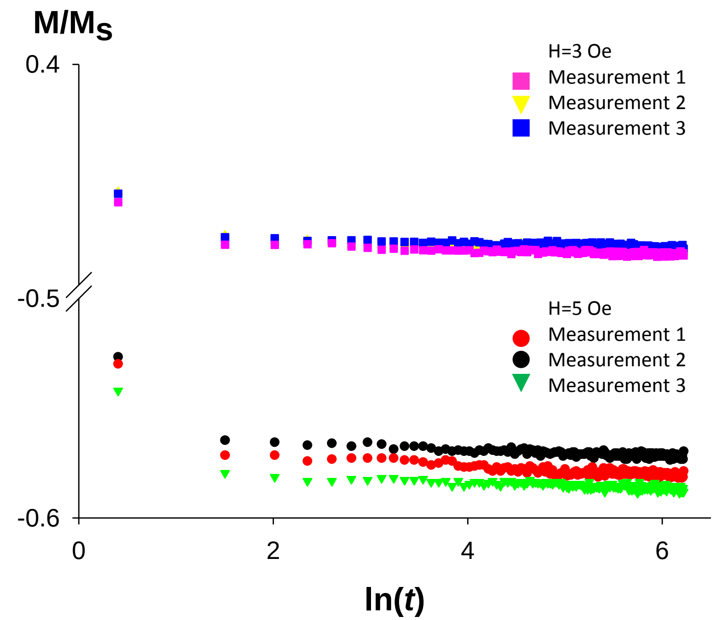
- Magnetic time dependence measurements have been taken over the switching region.

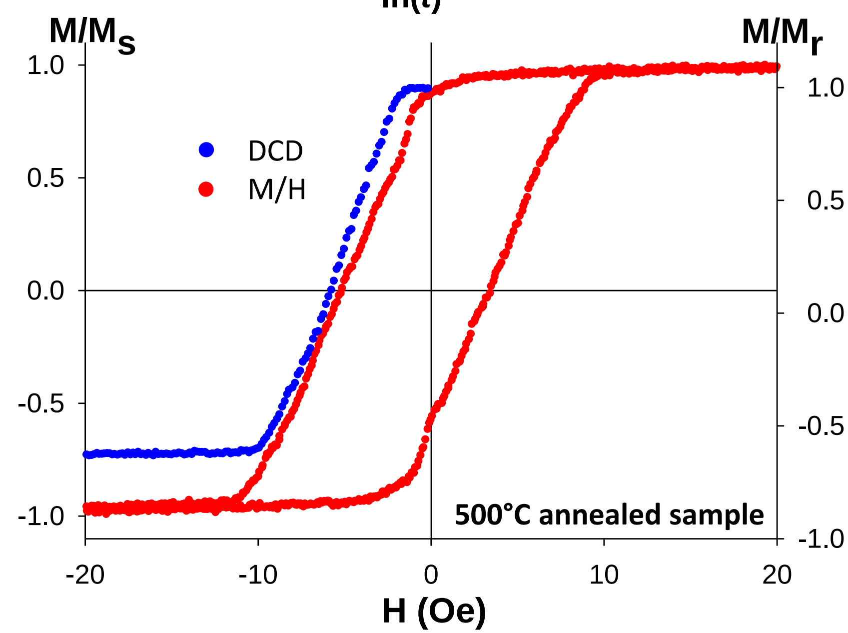
- These measurements are used to find a value for the magnetic viscosity $S_1(H)$ of each sample.

$$\frac{-dM(H)}{d\ln t} = S_0 + S_1(H)\ln t + (S_2(H)\ln t)^2 + ...$$

- DC demagnetised (DCD) remanence curves have also been taken for each sample.

- For a DCD curve the sample is saturated, then the remanent magnetisation (M_r) is measured at increasing values of negative field.





Fluctuation Field

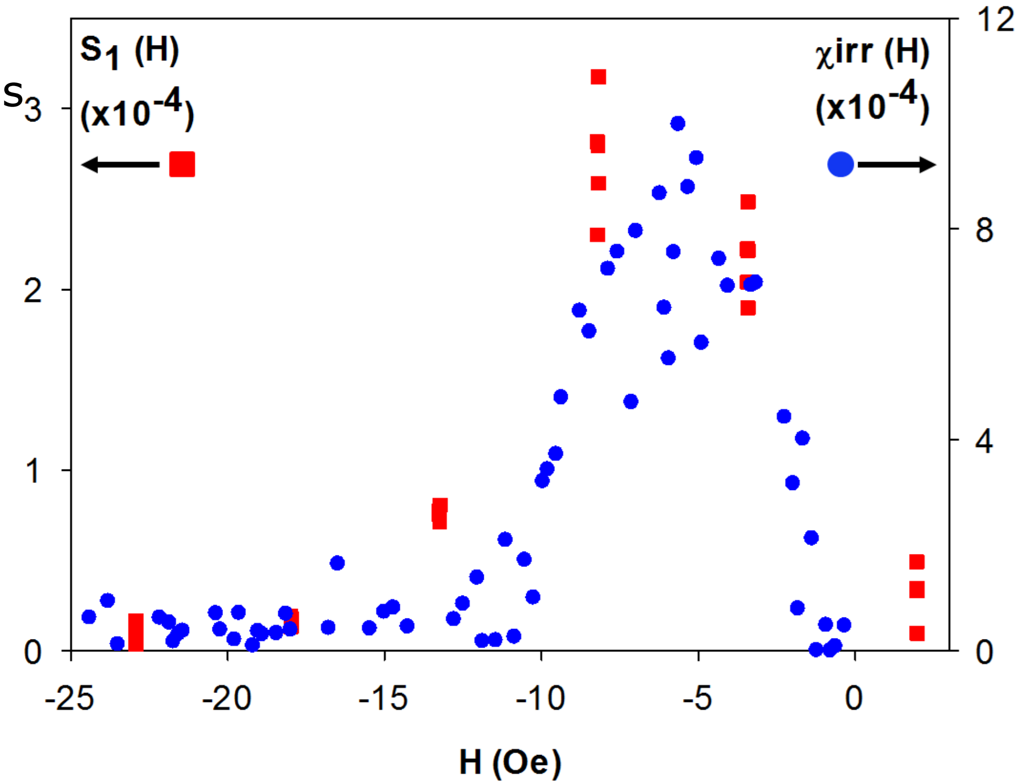
- The fluctuation field (H_f) is an imaginary field representing the effect of thermal energy.*

- The differential of the DCD curve gives the irreversible susceptibility (χ_{irr}).

$$\chi_{irr}(H) = \frac{d(DCD(H))}{dH}$$

- This can be combined with the value for $S_1(H)$ to give the fluctuation field (H_f).

$$H_f = \frac{S_1(H)}{\chi_{irr}(H)}$$

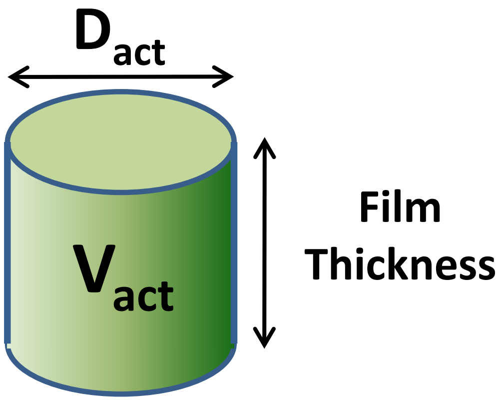


* L. Néel, *Ann. Geophys.* **5**, 99 (1949).

Estimated Activation Volumes

- H_f then gives rise to the concept of the activation volume (V_{act}) : *

$$V_{act} = \frac{k_B T}{M_s H_f}$$



- V_{act} is defined as the smallest volume that reverses in a single step.

- V_{act} is a relative measure because the value of M_s is unsure.

Annealing Condition	M_s (±0.1 emu/cc x10^4)	V_{act} (±0.5x10^{-17}cm^3)	D_{act} (±0.5 nm)	H_c (±0.1 Oe)
As – deposited	4.4	4.0	5.0	2.9
400˚C	5.2	1.6	3.2	1.7
500˚C	4.5	4.5	5.3	4.5
600˚C	4.8	4.6	5.4	7.2

* E. P. Wohlfarth, *J. Phys. F: Met. Phys.* **14**, 155 (1984).

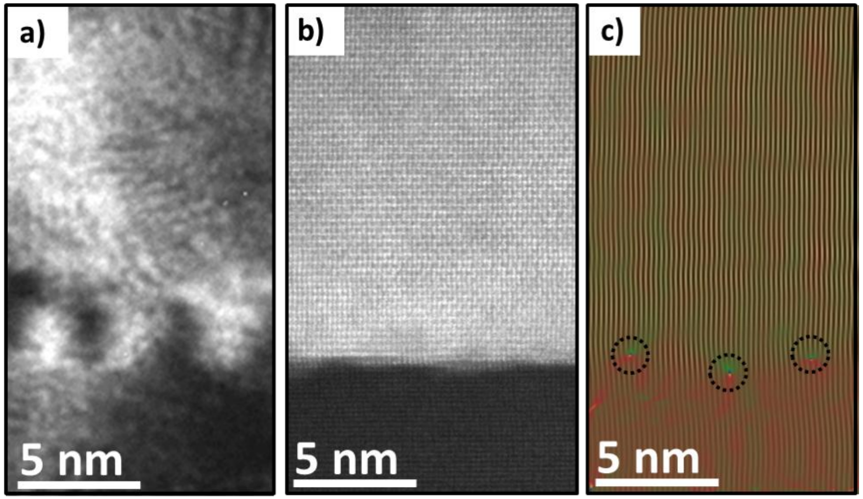
Activation Volume in Epitaxial Co_2FeSi

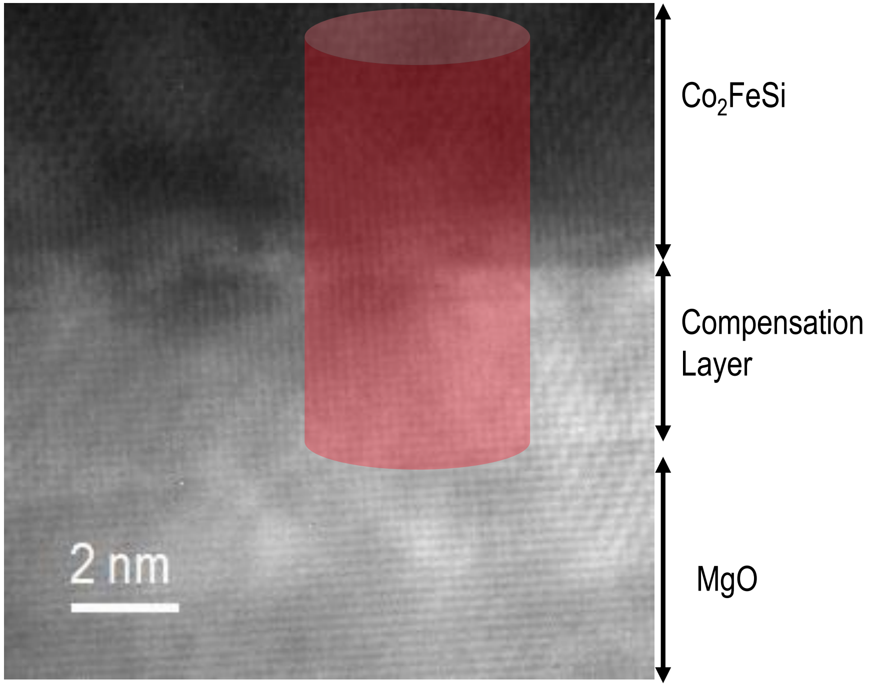
- Activation volume was estimated to be ~ 4.0 nm.

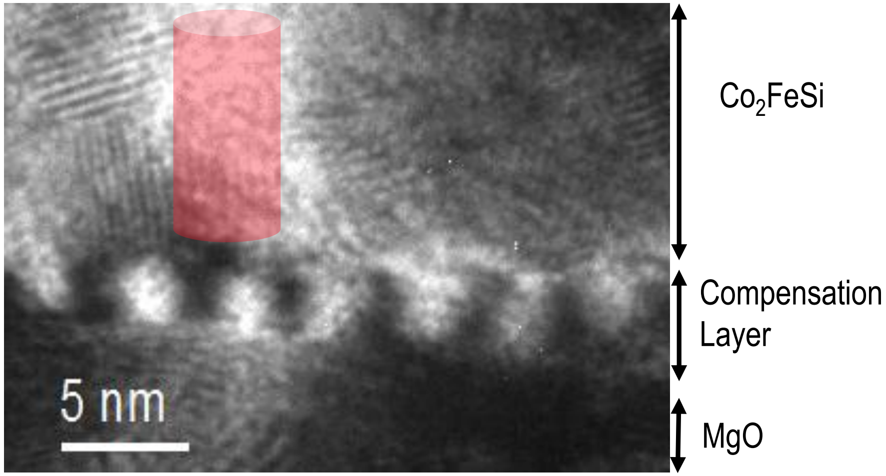
- A lattice mismatch of 4.5 % to be compensated between MgO and Co_2FeSi.

- This compensation layer (periodic contrast) can be seen in the contrast change in the TEM image due to an increase in lattice spacing through this layer.

- This contrast change is due to compensation through the Co_2FeSi missing entire MgO planes to improve the epitaxy.







* A. Hirohata *et al.*, *Appl. Phys. A* **111**, 423 (2013).

Pinned / Unpinned Domain Wall

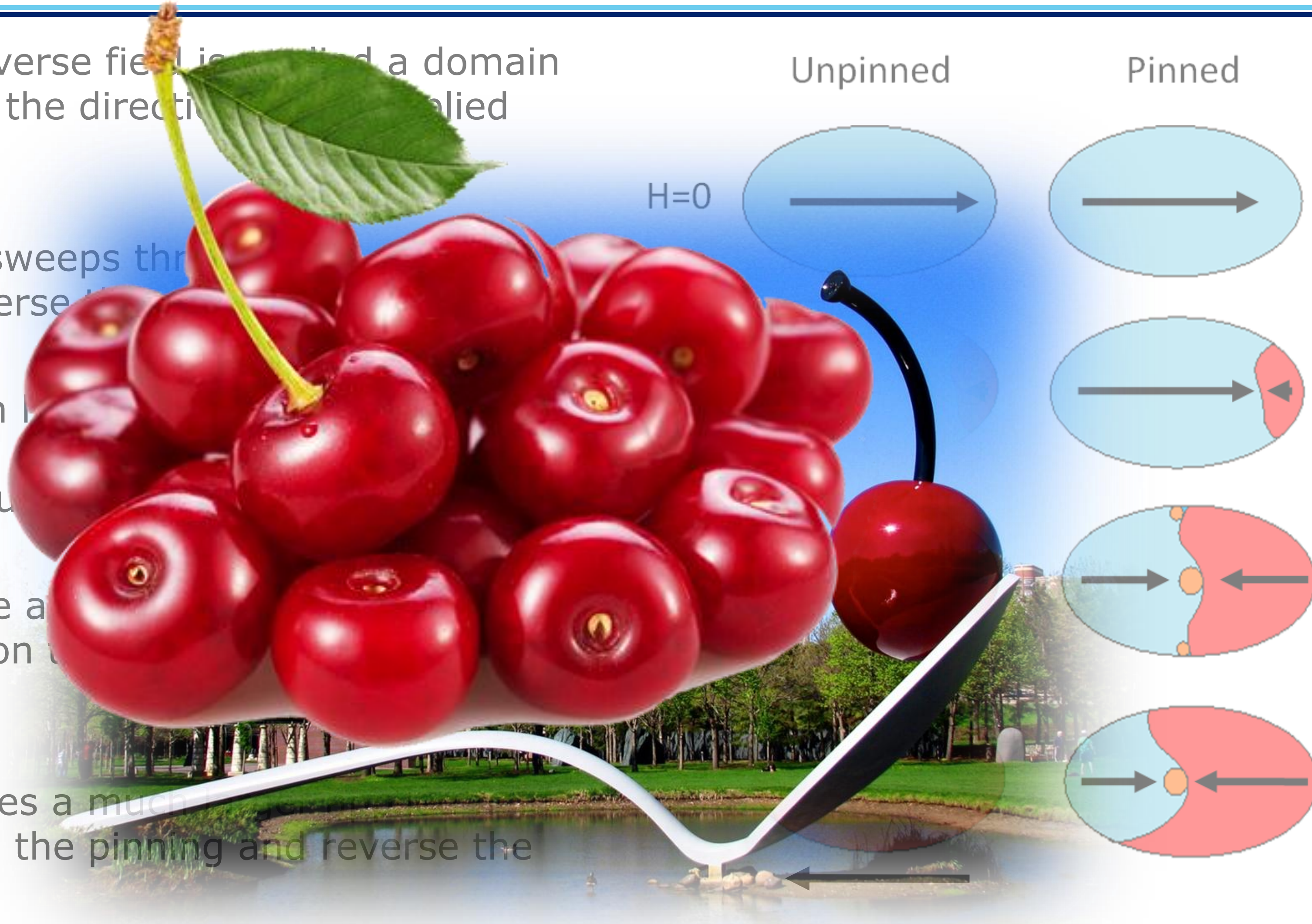
- When a reverse fie[...] a domain nucleates in the directi[...]lied field.

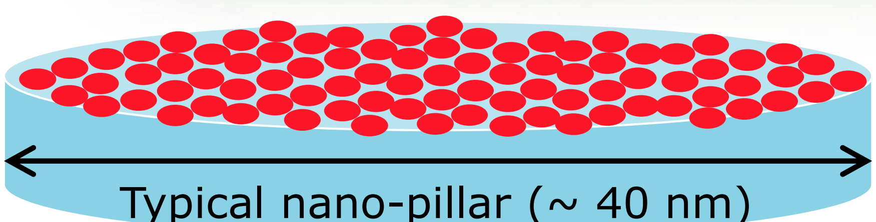
- This then sweeps thr[...] order to reverse [...]

- If the grain [...] ordering the [...] quickly and u[...]

- If there are a[...] contamination [...] pinned.

- It then takes a much [...] to overcome the pinning and reverse the entire grain.





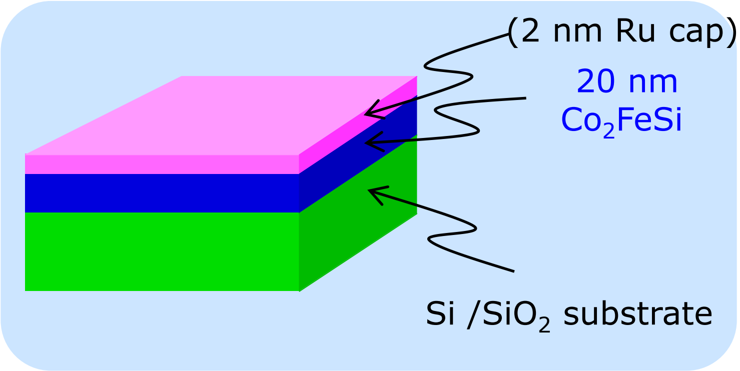
~ 25 pinning sites (epitaxial)

* J. Sagar *et al.*, *Appl. Phys. Lett.* **101**, 102410 (2013).

Heusler-Alloy Film Growth

Sputter film deposition : *

- Controlled plasma HiTUS sputtering system
- Optimised target composition (*e.g.*, $Co_{1.748}Mn_{1.118}Si_{1.134}$)
- Base pressure : < 3.0×10^{-5} Pa
- MgO (001) substrate cleaning :
  acetone bath for 10 min. +
  in situ heat treatment at 573 K for 20 min.
- Plasma :
  RF field at 3.0×10^{-1} Pa Ar pressure
  DC bias steering from -250 to -990 V to change the grain size
- Annealing at 760 K for 3 h (1st anneal)
  followed by further annealing at 760 K for another 3 h (2nd anneal)
  additional annealing at 760 K for 3 h (3rd anneal)



Magnetic / structural measurements :

- Princeton AGFM Model 2900
- ADE Model 10 VSM
- JEOL JEM-2011 TEM
- JEOL JEM-2200FS HR-(S)TEM

* A. Hirohata *et al.*, *Appl. Phys. Lett.* 95, 252506 (2009).

Activation Volume in Polycrystalline Co_2FeSi

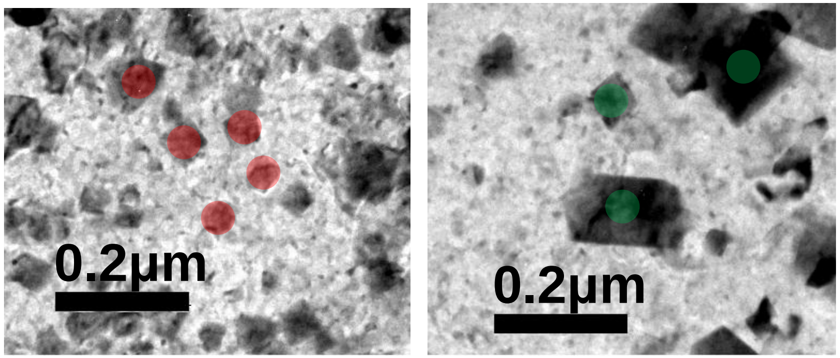
- The activation volume has been shown to be bias voltage independent (~ 40 nm), but varies with annealing time.

- The physical grain volume is shown to increase with bias voltage and vary with annealing time.

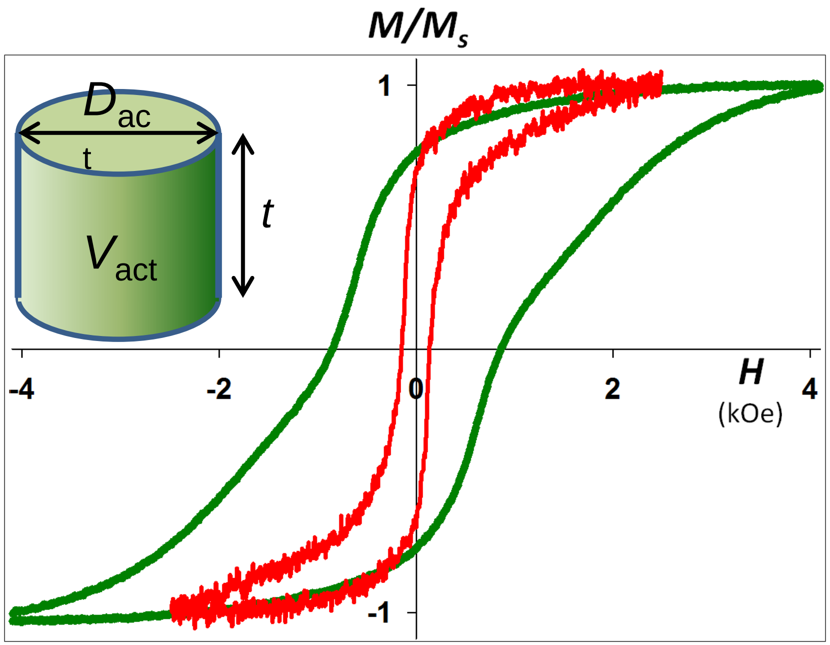
- For films deposited at higher bias voltages the activation volume was 40% of the physical volume the particles are therefore multi-domain.

- Polycrystalline films can offer a "pinning-site-free" nano-pillar.

250V **3 Hour Anneal** | **750V** **6 Hour Anneal**



= D_{act} 250V   = D_{act} 750V

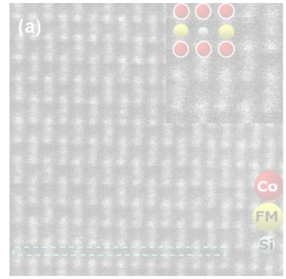


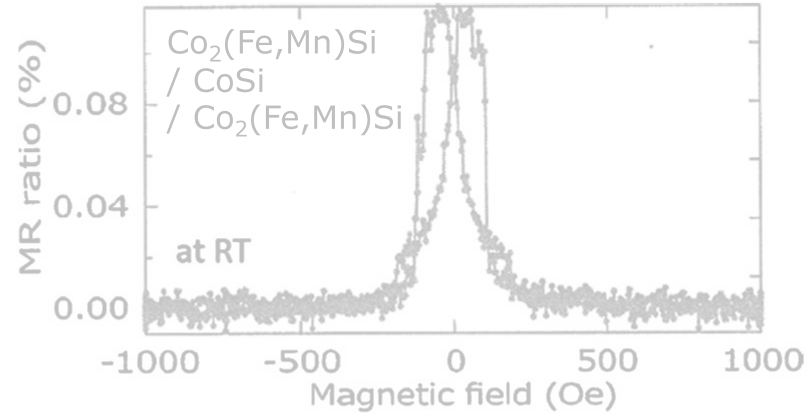
* J. Sagar *et al.*, *IEEE Trans. Magn.* 47, 2440 (2011).

Possible Solutions

Smoothing the interfaces :

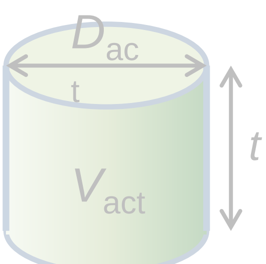
- Optimisation of the non-magnetic spacer
  - → Ag
- Atomically sharp interface achieved
  - → GMR ratio : ~ 15 %

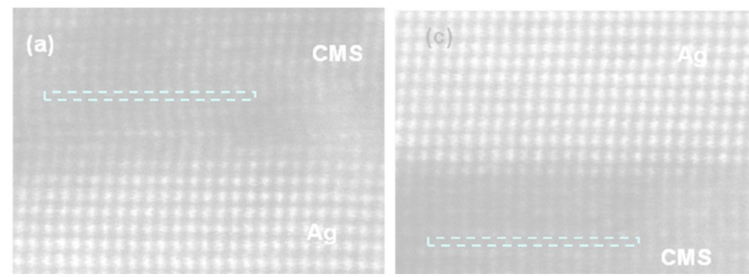


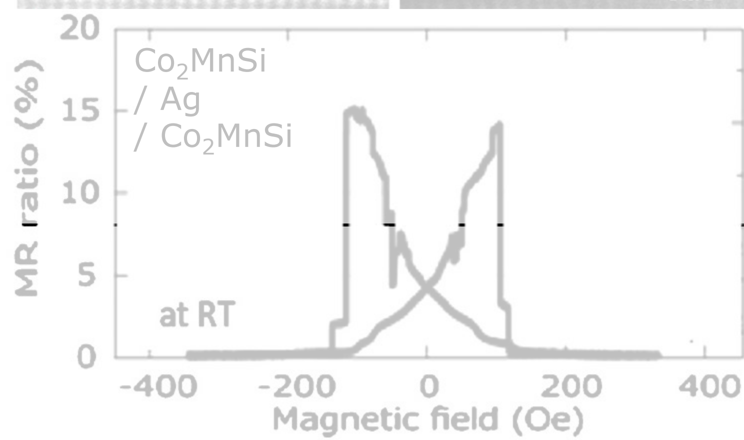


Elimination of minor domains :

- Maximisation of activation volume
  - → smallest volume that reverses in a single step







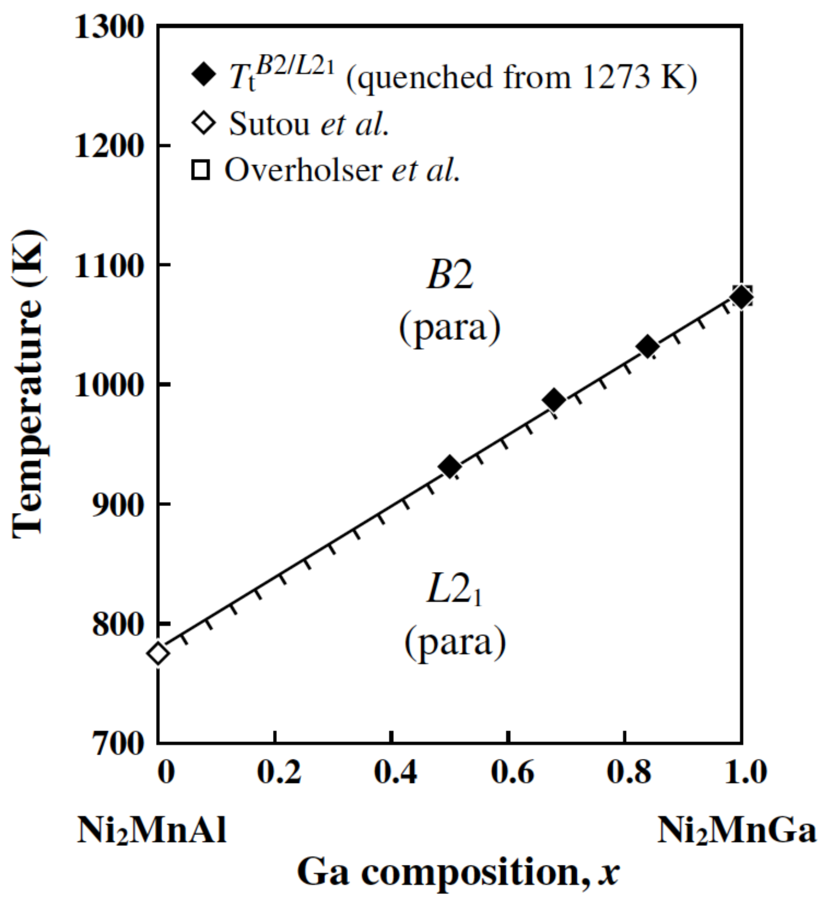
Minimisation of intermixing / deformation :

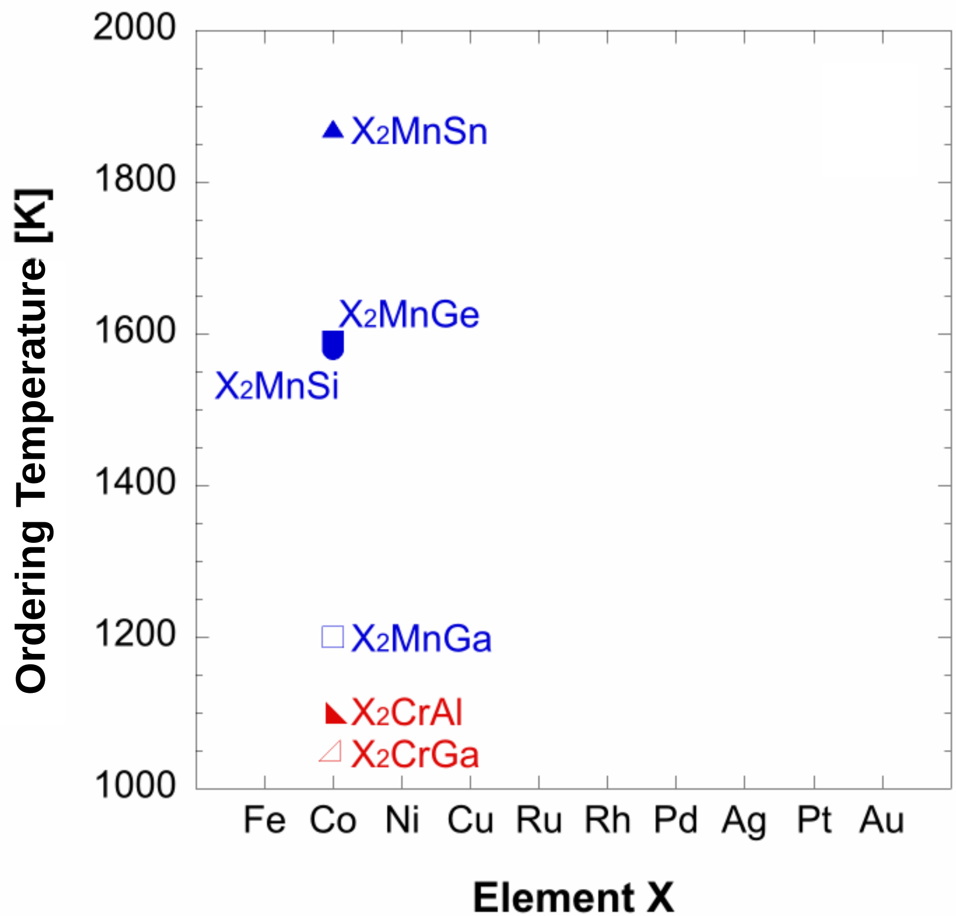
- Low-temperature annealing for Heusler alloy films
  - → *in situ* TEM observation

* V. K. Lazarov *et al.*, *Appl. Phys. Lett.* 98, 242508 (2011).

Ordering Temperature for Heusler Alloys

For example, $Ni_2Mn(Ga,Al)$: *





* H. Ishikawa *et al.*, *Acta Mater.* **56**, 4789 (2008);
** A. Hirohata *et al.*, *Heusler Alloys* (Springer, in press).

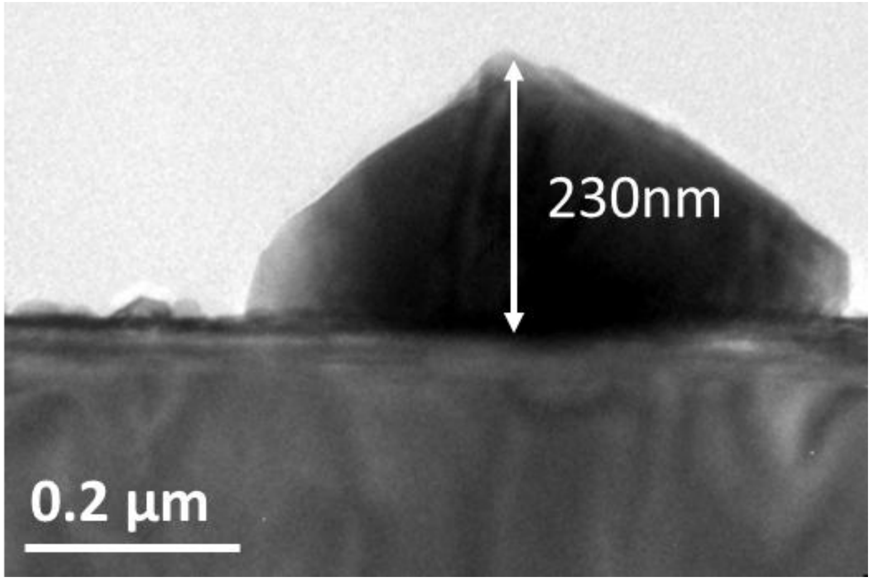
Deformation of Polycrystalline Co_2FeSi

- After annealing at 500 °C for 6 h, a 20 nm thick Co_2FeSi film crystallises 3-dimensionally.

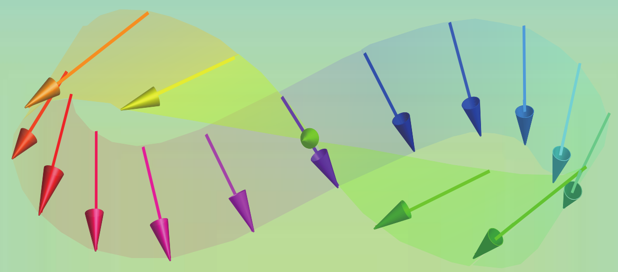
- This forms ~ 230 nm high grain.

- This induces the discontinuity of the Co_2FeSi films.

- Lower-temperature annealing with shorter period is necessary to minimise the deformation.



Low-Temperature Crystallisation of Heusler Alloy Films



J. Sagar,[1] L. R. Fleet,[1] M. Walsh,[1,2] K. Yoshida,[3,4] V. K. Lazarov,[1]
E. D. Boyes,[1,2] T. Nakayama [4] and A. Hirohata [1]

1 The University of York

2 York JEOL Nanocentre

3 Nagoya University

4 JFCC

5 長岡技術科学大学 Nagaoka University of Technology

JST さきがけ PRESTO

EPSRC Engineering and Physical Sciences Research Council
EP/H026126/1
EP/M02458X/1

Grain Crystallisation Process

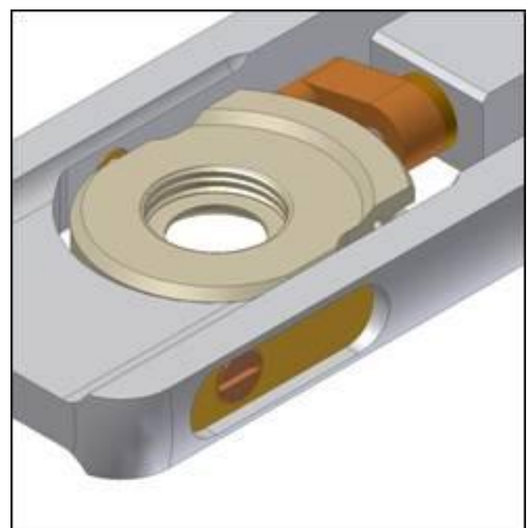
In situ TEM observation :

JEOL JEM-2200FS :

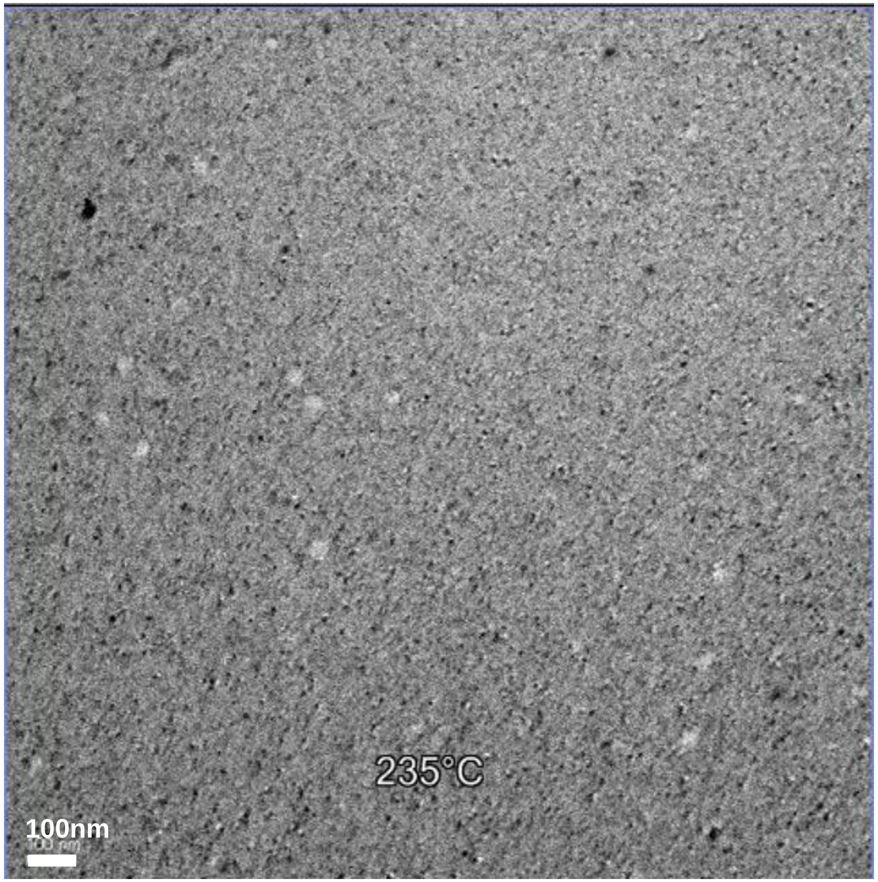
- Double Cs correction
- Gas introduction to sample space
- Gatan heating sample stage (< 700°C)

Heusler-alloy films :

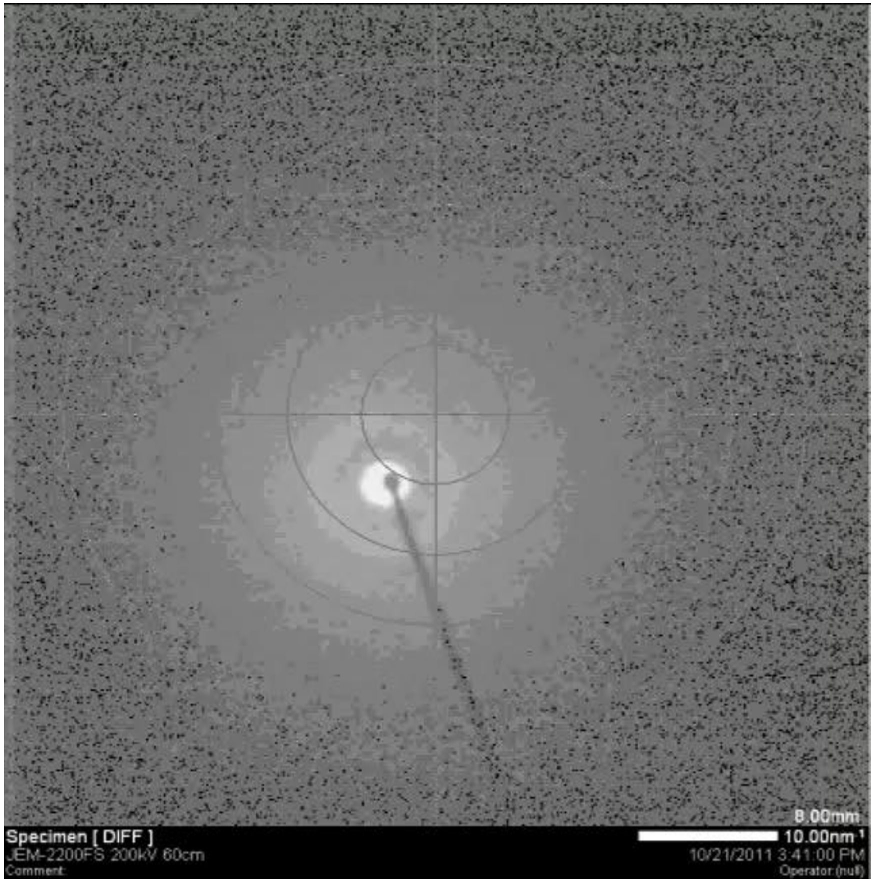
- 20 nm Co_2FeSi / 2 nm Ru
- Grown on SiN TEM grids
- Continuous movie (Camtasia studio)
- Detailed HRTEM / diffractograms



In Situ Crystallisation Process



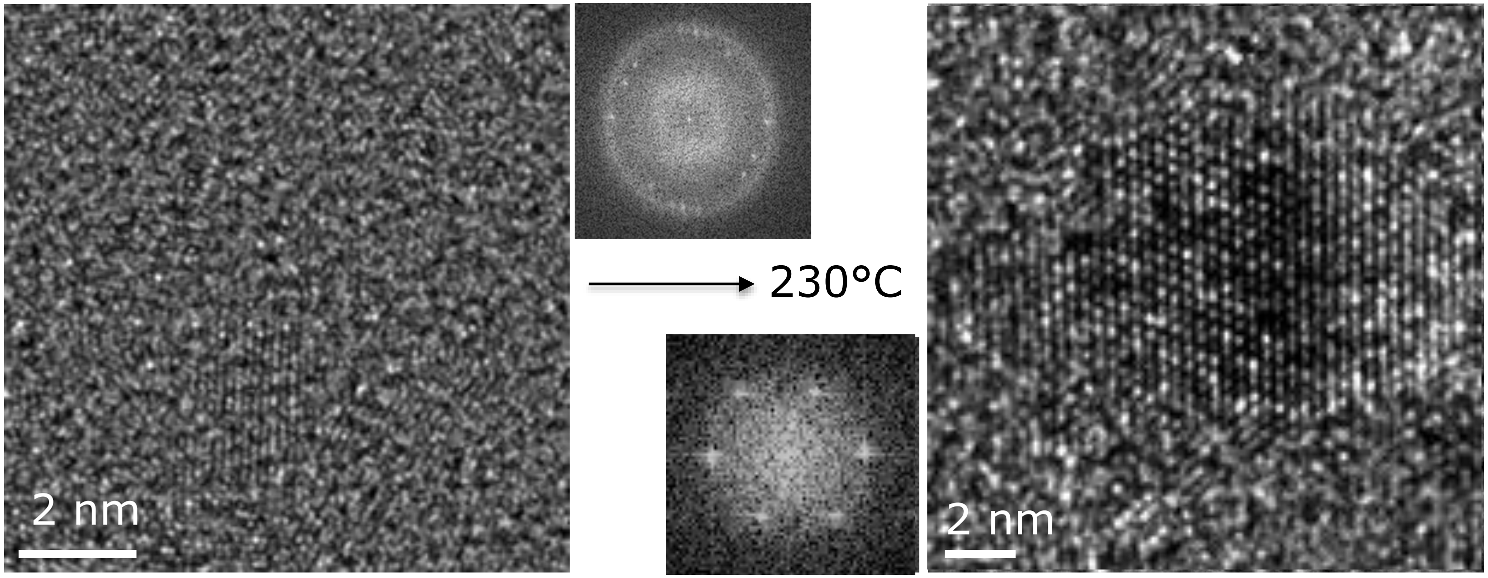
Bright Field TEM
(235°C for 3 hours)

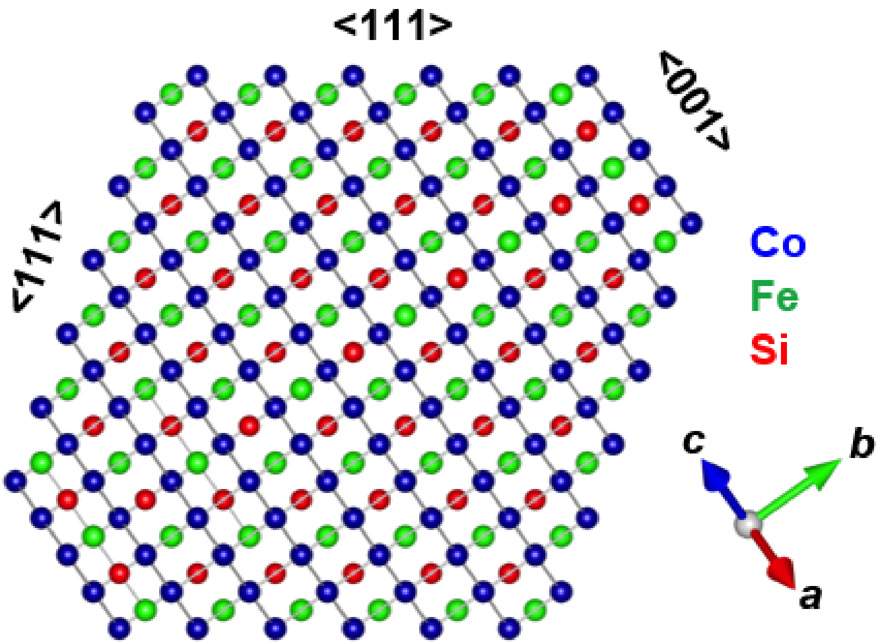


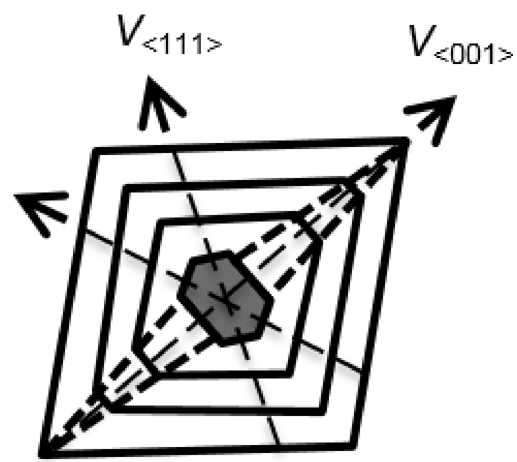
Electron diffraction pattern
(235°C for 3 hours)

Structural Analysis of Individual Grains

Initial grain nucleation :



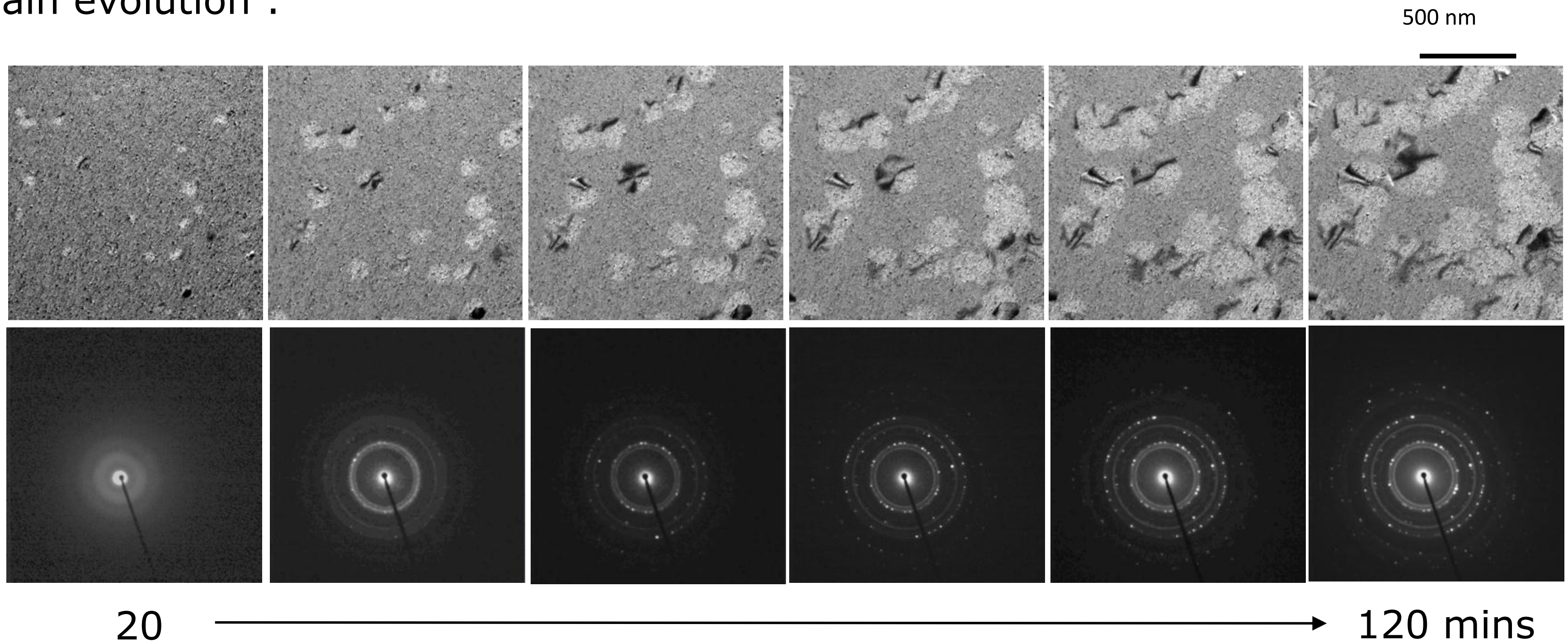


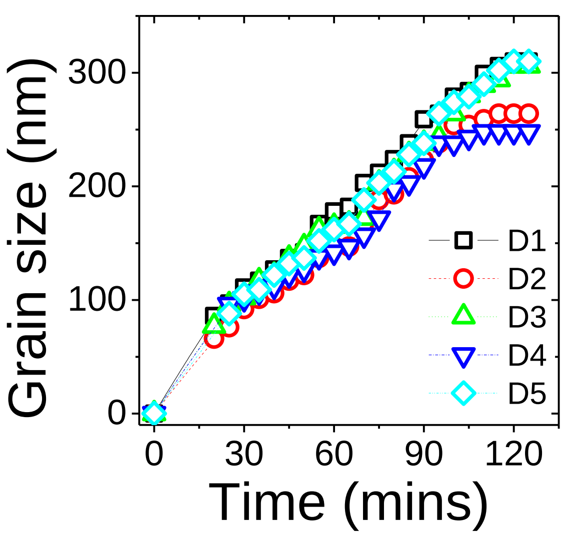


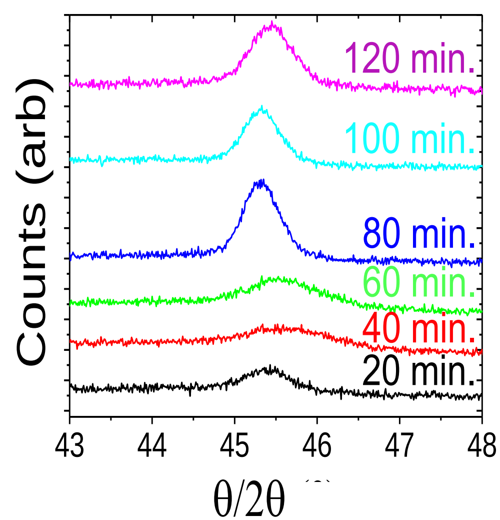
* J. Sagar *et al.*, *Appl. Phys. Lett.* **105**, 032401 (2014).

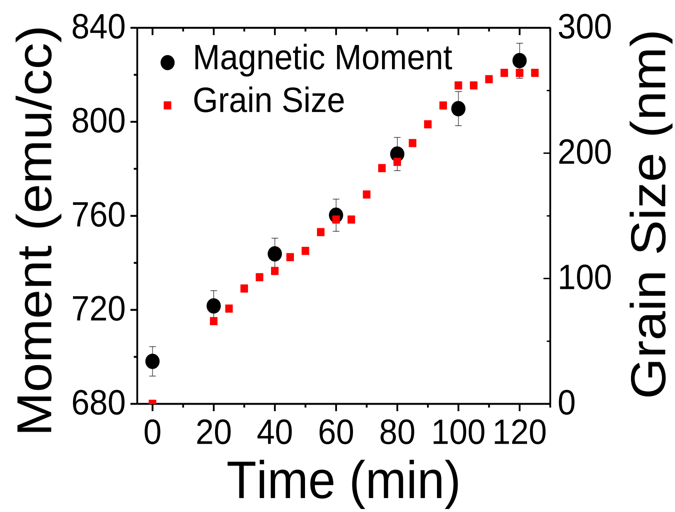
Structural Analysis of Individual Grains

Grain evolution :

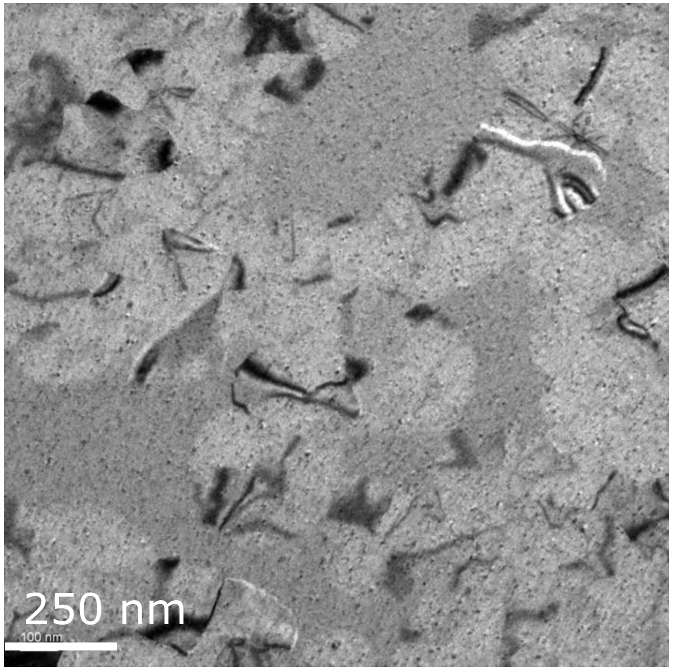


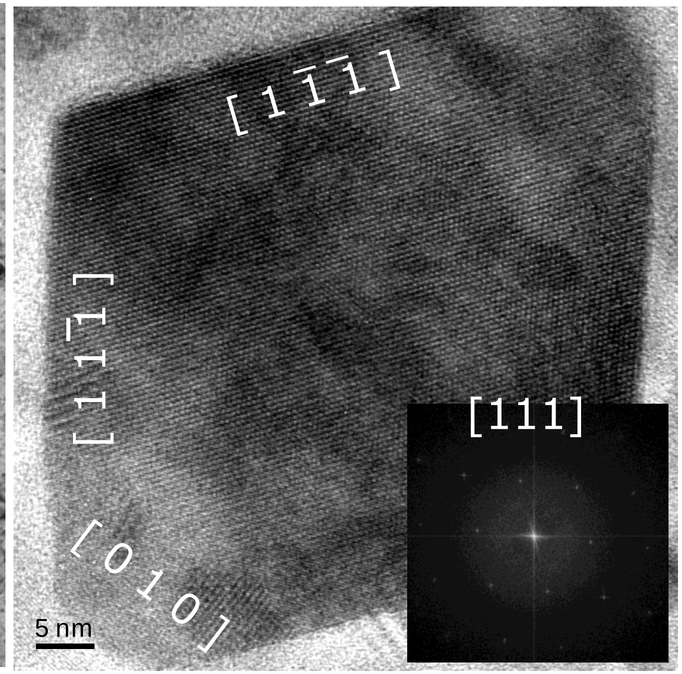


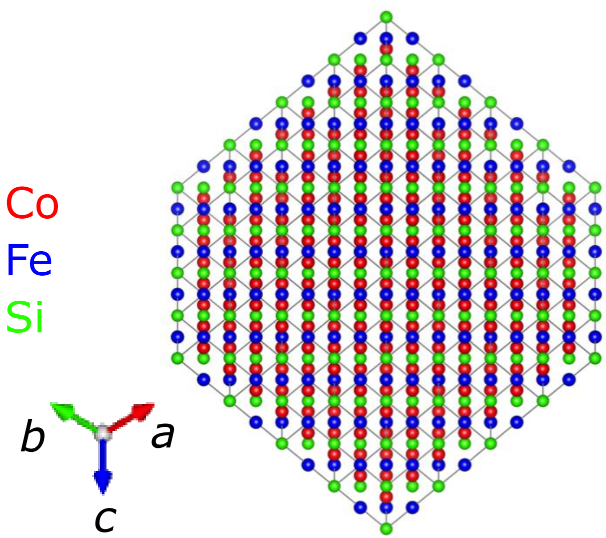


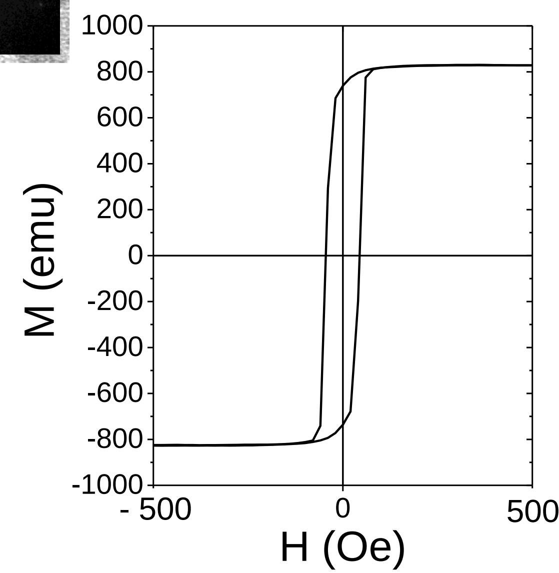


Grain crystalline orientation :









* A. Hirohata *et al.*, *British Patent*, GB1402399.8.

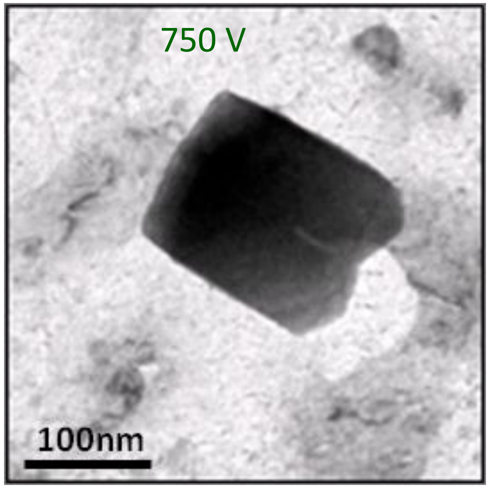
Higher Temperature Annealing

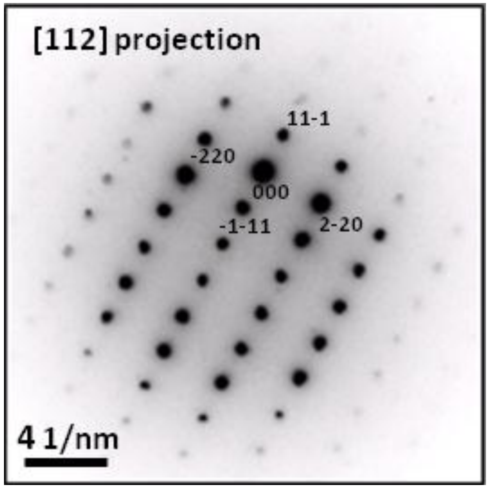
Structural analysis on Co_2FeSi grains using HRTEM :

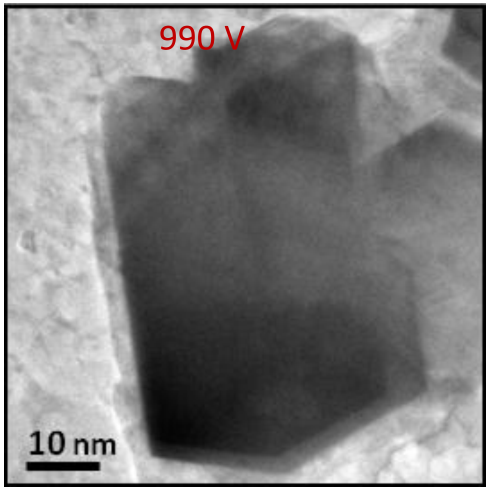
- The analysis was performed on the 6-hour annealed sample with the maximum grain size.

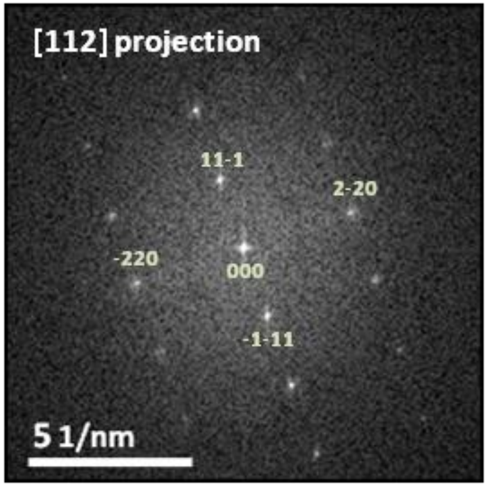
- The grains were analysed using SAED patterns and digital diffractograms of HRTEM images.

- Each of the grains analysed showed a very well ordered structure, lying predominantly along the [112] orientation, possibly in the $L2_1$ phase.







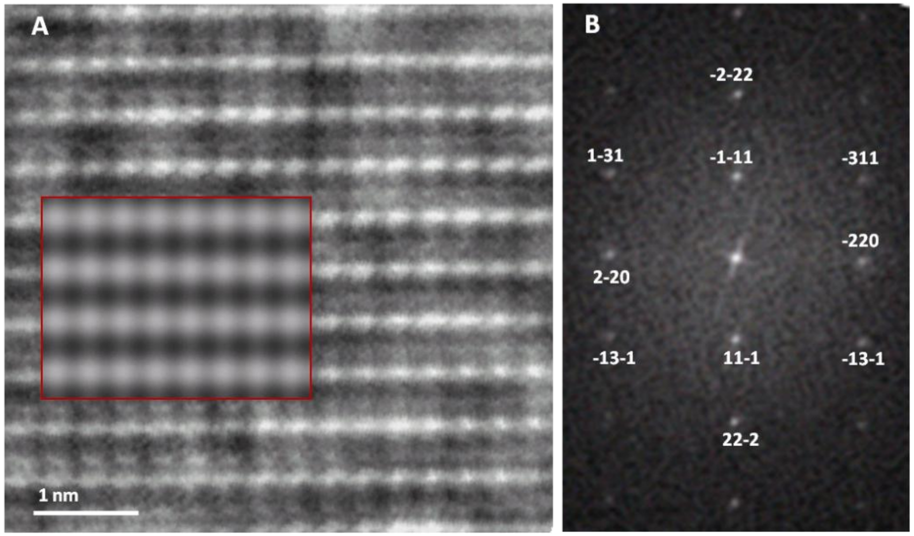


* L. R. Fleet *et al.*, *J. Phys. D: Appl. Phys.* **45**, 032001(FTC) (2012).

Layer-by-Layer Crystallisation

Cross-sectional analysis on Co_2FeSi films using HRTEM :

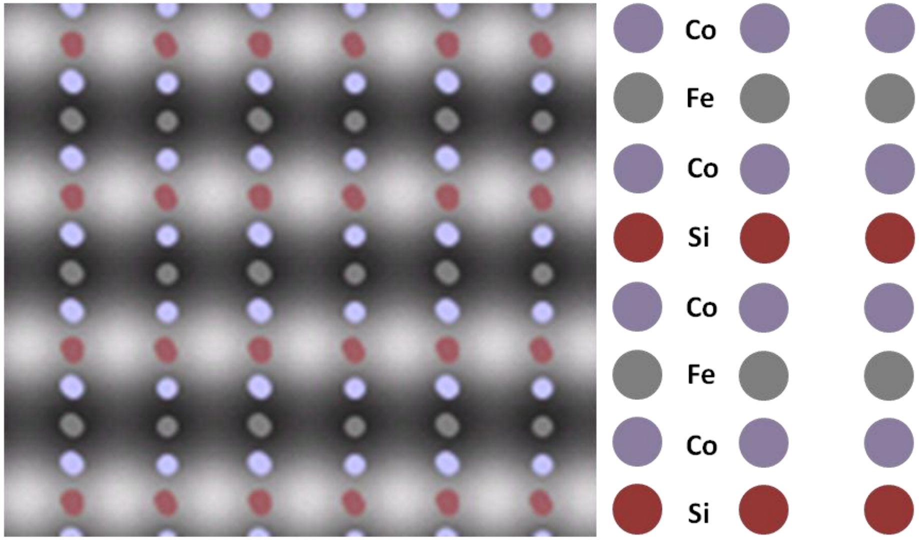
V_B = 990 V after 9 h annealing



- Single-nanocrystalline grains with the $L2_1$ phase were observed along the [112] axis.

- Image simulations were produced using the multislice method in the electron microscopy software JEMS. *

- The grains were assumed to be in the $L2_1$ phase, orientated along the [112] zone axis.



- The simulations were found to match with the experimental HRTEM images.

- This implies the grains are in the $L2_1$ phase and crystallised in a layer-by-layer growth mode.

* P. Stadelmann, http://cimewww.epfl.ch/people/Stadelmann/jemsWebSite/jems.html.

Summary on Ferromagnetic Heusler Alloys

- Small grains of ~ 10 nm begin to form at around 230°C. These grains continue to grow up to ~ 200 nm in size when held at 230°C for 3 hours.

- The lattice constant was estimated to be 0.565 nm (expected for $L2_1$ ordering).

- Further annealing does not appear to cause any significant change in the films but does effect the structure of the grains with striping occurring after annealing over 500°C.

- Magnetic moments gave 80 % of the theoretical maximum value.

  → Evidence for the presence of the $L2_1$ phase.

- HRTEM images and SAED patterns show ordered grains lying in the (110) orientation.

  → Grains crystallising in a layer-by-layer mode.

Heusler Alloy Replacement for Iridium

HARFIR

A. Hirohata,[1] G. Reiss,[2] U. Nowak,[3] L. Sznyogh,[4]
K. Takanashi [5] and K. Ono [6]

1 The University of York
2 Universität Bielefeld
3 Universität Konstanz
4
5 TOHOKU UNIVERSITY
6 KEK

NMP3-SL-2013-604398 SEVENTH FRAMEWORK PROGRAMME JST *SCICORP-2013*

The Scarcest Material

- Melting point : > 3,000° C
  → Very stable
- Almost no applications previously

- World supply : ~ 5.8 t / yr
  → 87 % from South Africa
- 1 ~ 2 % in Pt and Rh ore

- The scarecest element
  → 4 × 10^{-4} ppm
  Comparisons Nd : 33 ppm
  Li : 17 ppm
  Dy : 6.2 ppm
  Pt : 3.7 × 10^{-3} ppm
  Au : 3.1 × 10^{-3} ppm
  Ru : 1 × 10^{-3} ppm

* http://www.atheistfrontier.com/people/dmitri-mendeleyev/periodic-table-of-the-elements-for-kids.jpg

Project Objectives

Aim

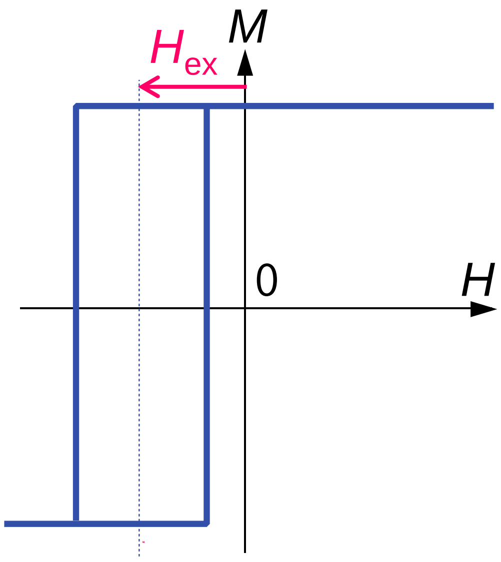
IrMn alloy used in GMR / TMR junctions
→ Antiferromagnetic Heusler alloys with common elements

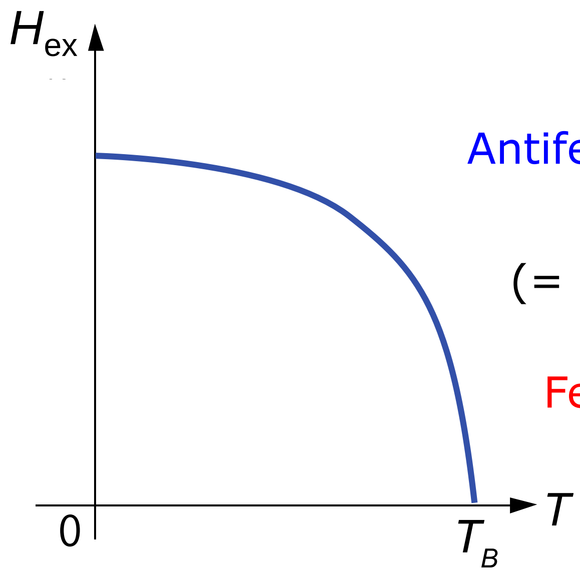
Objectives

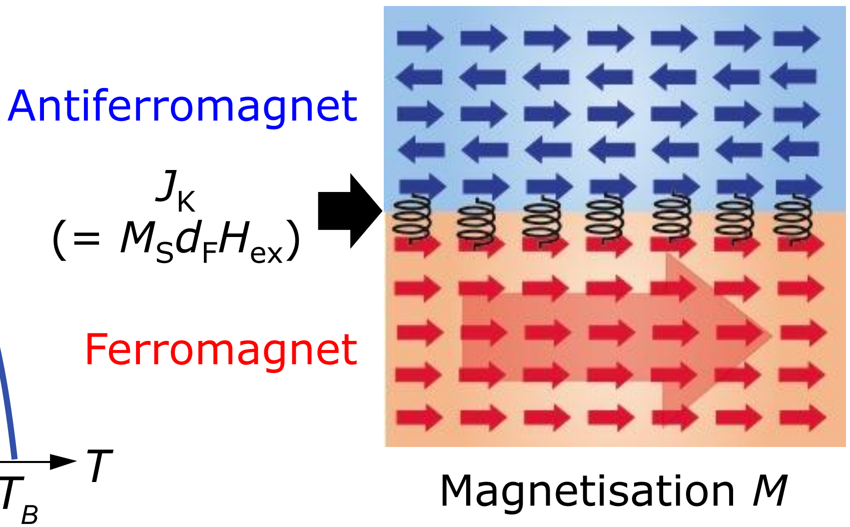
Exchange bias : H_{ex} > 1 kOe (J_K > 1 erg/cm^2)

Blocking temperature : T_B > 300 K

Distribution of the blocking temperature : σ_{TB} < 0.3







Summary on Antiferromagnetic Heusler Alloys

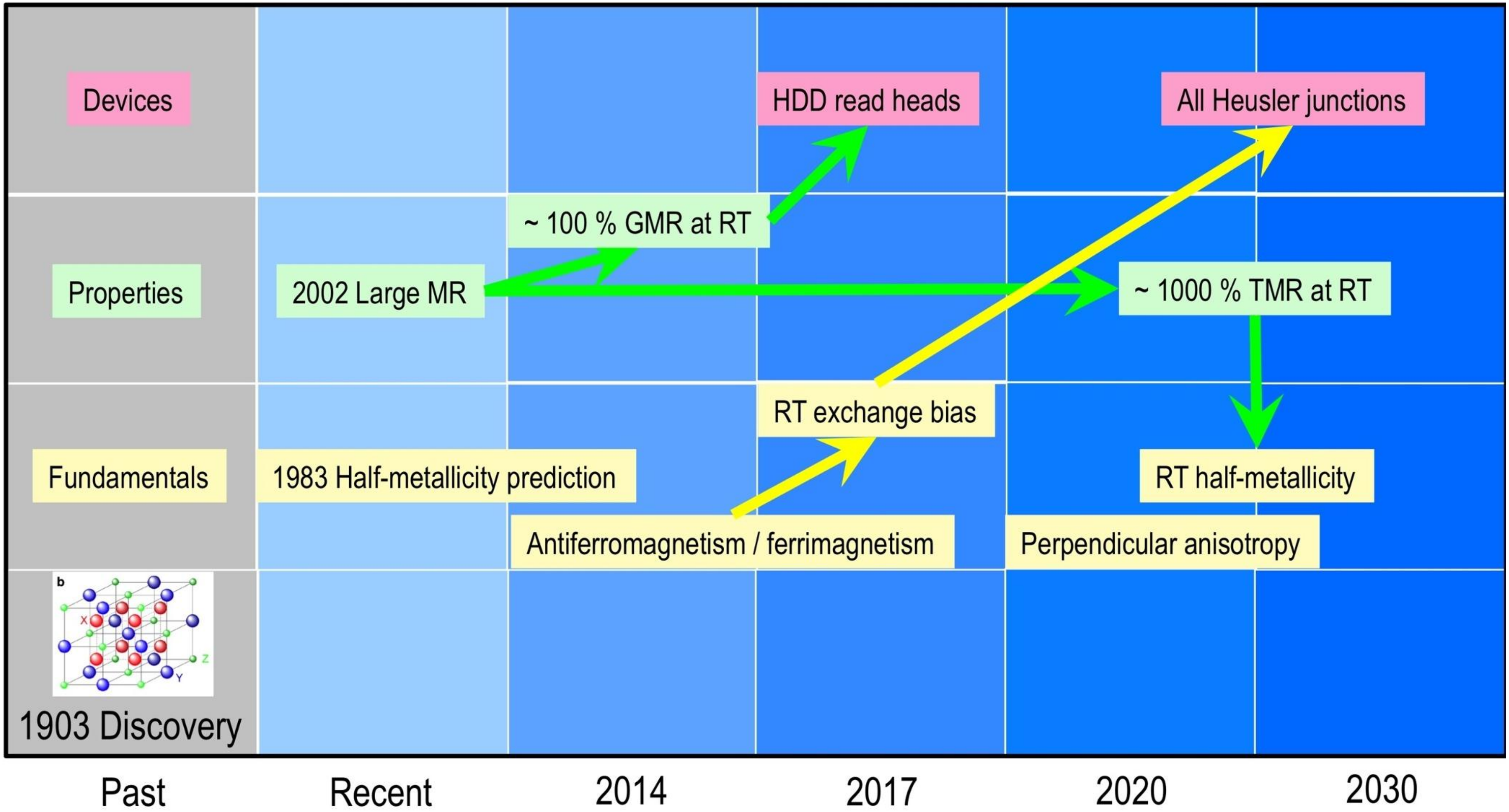
- ❑ Antiferromagnetic Heusler alloys have been successfully grown.
- ❑ Exchange bias has been observed at low temperature.
- ❑ Further optimisation on the crystallisation is required to achieve antiferromagnetism at room temperature.

  → Such a layer is ideal for next-generation junctions.

Polycrystalline Heusler-alloy films can

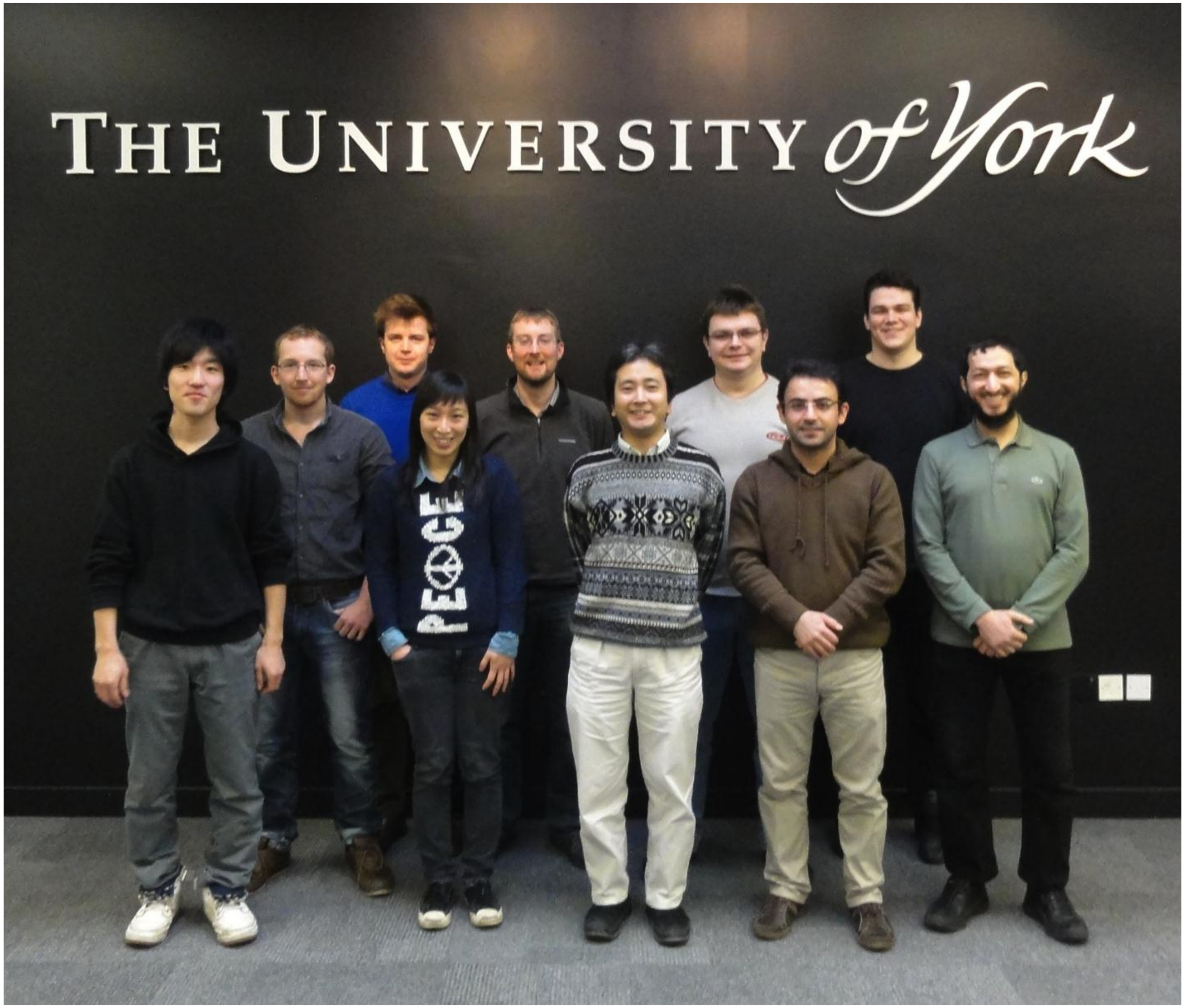
- form a junction to be reversed in a single step.
- crystallise at below 300° C compatible with the CMOS technology.
- exhibit exchange bias against a ferromagnetic layer.

Roadmap on Heusler Alloys



* A. Hirohata *et al.*, *IEEE Trans. Magn.* **51**, 07160747 (2015).

Other Group Activities



Special thanks to …

& their students

& York colleagues

13th Joint MMM-Intermag Conference

January 11-15, 2016

San Diego, California

Conference Chair:

Bruce Gurney (*Bruce_Gurney@ieee.org*)

Program Co-Chairs:

Katayun Barmak (*kb2612@columbia.edu*)

Atsufumi Hirohata (*atsufumi.hirohata@york.ac.uk*)

http://www.magnetism.org/

Abstract submission: 7 August 2015

IEEE MAGNETICS

AIP Publishing

Thank you very much for your attention.

Atsufumi Hirohata

Department of Electronics
University of York
+44 (0)1904 32 3245
atsufumi.hirohata@york.ac.uk

york spintronics

JST さきがけ PRESTO

THE ROYAL SOCIETY

EPSRC
Engineering and Physical Sciences Research Council

SEVENTH FRAMEWORK PROGRAMME

SEAGATE