S2.A 3781

Cross-Sectional TEM Imaging of NiCrMnSi and CoFe:N Alloys for Magnetic Tunnel Junctions

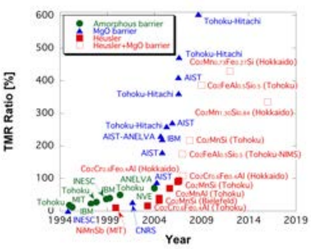
K. Elphick,[1] Y. Onodera,[2] T. Tsuchiya,[2] T. Ichinose,[2] S. Mizukami,[2] K. Yoshida,[3] Y. Nagai[3] and A. Hirohata[1]

[1] *Department of Electronic Engineering, University of York, York YO10 5DD, United Kingdom*

[2] *WPI- Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

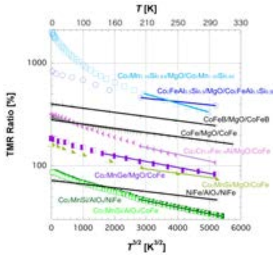
[3] *Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

Recent development of TMR ratios [1]



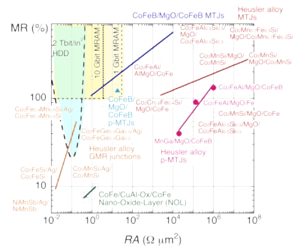
- Amorphous barriers achieved TMR ratios of < 100% at room temperature (RT).
- Epitaxial barriers improved TMR ratios up to ~ 600% at RT due to the coherent tunnelling.
- Half-metallic Heusler alloys show similar TMR ratios at RT.
- In this study, we investigated Heusler alloys and nitrides for magnetic tunnel junctions (MTJs).

Temperature dependence of TMR ratios [1]



- Amorphous barriers follow the empirical law of $T^{3/2}$.
- Epitaxial barriers show faster decrease than the empirical law of $T^{3/2}$.

Targets of TMR ratios [2]



- MTJs require further reduction in *RA*.

→ Thinner tunnel barrier

→ Smaller temperature dependence

[1] A. Hirohata *et al.*, *J. Magn. Magn. Mater.* **509**, 166711 (2020).

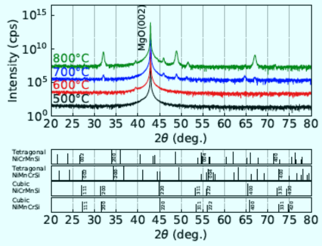
[2] A. Hirohata *et al.*, *Materials* **11**, 105 (2018).

Samples growth by ultrahigh vacuum sputtering

The CoFe:N and NiCrMnSi samples were sputtered using ultrahigh vacuum (UHV) magnetron sputtering.

The structures of the samples are as follows (thickness in nm) :

(i) MgO(001) sub.//NiCrMnSi (100)/Ta (3) [3]

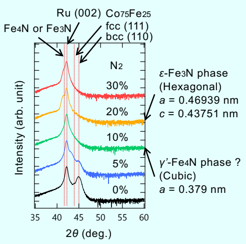


The NiCrMnSi samples were in situ annealed at T_a = 500 and 700ºC before the Ta layer was deposited.

(ii) Si sub.//Ta (5)/Ru (10)/$Co_{75}Fe_{25}$:N (15)/Ta (5) [4]

- N_2 10% (0.1 Pa N_2 + 0.9 Pa Ar)
- N_2 20% (0.2 Pa N_2 + 0.8 Pa Ar)

Deposition temperature was 250ºC.



[3] Y. Onodera *et al.*, *Jpn. J. Appl. Phys.* **59**, 073003 (2020).

[4] T. Ichinose *et al.*, *ACS Appl. Mater. Interfaces* **1**, 2220 (2019).

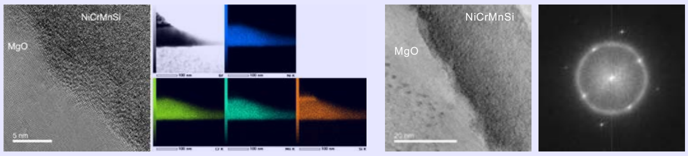
NiCrMnSi

High resolution transmission electron microscopy (HR-TEM) and energy dispersive X-ray spectroscopy (EDX) mapping :

The sample grown at T_a = 500ºC shows atomic mixing at the MgO(001)/NiCrMnSi interface up to 5 monolayers (MLs).

→ Polycrystalline nature, even though the EDX mapping proves all the constituent elements are distributed homogeneously.

→ The growth temperature of 500ºC is not enough for the NiCrMnSi crystallisation.



The sample grown at T_a = 700ºC is epitaxially grown with the grain size of 5~10 nm with interfacial mixing of ~ 20 MLs.

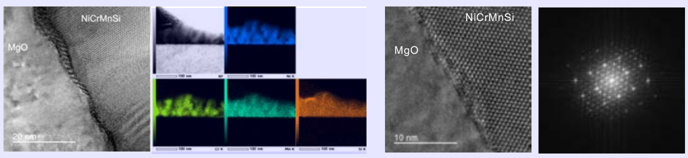
→ NiCrMnSi crystallisation.

→ This agrees with the X-ray diffraction (XRD) signals measured on these devices.

→ The corresponding EDX map shows Ni and Cr segregations from the NiCrMnSi matrix, which agrees with XRD signals.

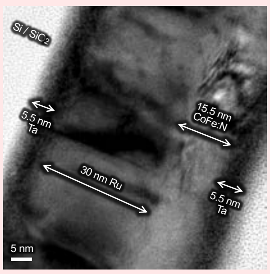
The corresponding transport properties of their MTJs show larger TMR ratios in the sample grown at T_a = 700ºC.

→ The interfacial smoothness of the MTJs rather than the compositions of the ferromagnetic layers.

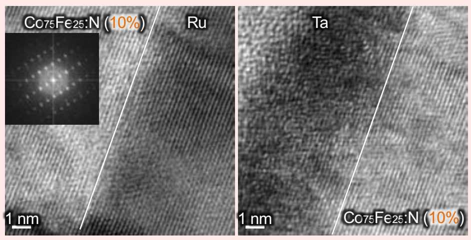


$Co_{75}Fe_{25}$:N

Interfacial roughness of ~ 0.3 nm at Ru / of $Co_{75}Fe_{25}$:N and ~ 1.0 nm at of $Co_{75}Fe_{25}$:N / Ta



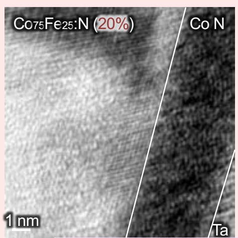
High resolution (1.5M magnification) TEM images of $Co_{75}Fe_{25}$:N (N_2 10%) :

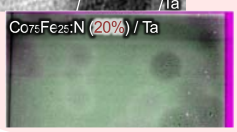


- Clear epitaxial growth of $Co_{75}Fe_{25}$:N is observed.
- No contaminations / phase segregations detected at $Co_{75}Fe_{25}$:N interfaces.

- Bottom Ru / $Co_{75}Fe_{25}$:N interface is observed by subtracting SEM images between 2.2 and 2.0 keV.
- Top $Co_{75}Fe_{25}$:N / Ta interface is observed by subtracting SEM images between 0.7 and 0.6 keV.

→ ~ 1 μm cobalt-nitride grains are formed.





High resolution (1.5M magnification) TEM images of $Co_{75}Fe_{25}$:N (N_2 20%) :

- Clear epitaxial growth of $Co_{75}Fe_{25}$:N is observed.
- ~ 3 nm thick Co-N layer is observed at $Co_{75}Fe_{25}$:N interfaces.
- Bottom Ru / $Co_{75}Fe_{25}$:N interface is observed by subtracting SEM images between 2.2 and 2.0 keV.
- Top $Co_{75}Fe_{25}$:N / Ta interface is observed by subtracting SEM images between 0.7 and 0.6 keV.

→ ~ 3 μm cobalt-nitride grains are formed.

Summary

- We fabricated NiCrMnSi and $Co_{75}Fe_{25}$:N films for MTJs.
- Polycrystalline NiCrMnSi grown at 500ºC.
- Epitaxial NiCrMnSi grown at 700ºC but with Ni and Cr segregation.
- Non-destructive imaging confirmed Co-N formation at the $Co_{75}Fe_{25}$:N / Ta interfaces.
- Co-N thickness increases with increasing N_2 partial pressure during sputtering.
- To avoid Co-N formation, a BN tunnelling barrier was tested, showing – 0.2% TMR ratio at room temperature. [4]
- Further optimisation is required for Heusler-alloy and nitride-based MTJs. [5]

[5] K. Elphick *et al.*, http://arxiv.org/abs/2010.04493.

The University of York

TOHOKU UNIVERSITY

EPSRC Engineering and Physical Sciences Research Council

JST CREST JEOL Solutions for Innovation