

This is a repository copy of *Fabrication of magnetic tunnel junctions with a metastable bcc Co₃Mn disordered alloy as a bottom electrode*.

White Rose Research Online URL for this paper:

<https://eprints.whiterose.ac.uk/148451/>

Version: Accepted Version

Article:

Kunimatsu, Kazuma, Tsuchiya, Tomoki, Elphick, Kelvin et al. (4 more authors) (2019) Fabrication of magnetic tunnel junctions with a metastable bcc Co₃Mn disordered alloy as a bottom electrode. Japanese Journal of Applied Physics. 080908. ISSN 1347-4065

<https://doi.org/10.7567/1347-4065/ab2f96>

Reuse

Items deposited in White Rose Research Online are protected by copyright, with all rights reserved unless indicated otherwise. They may be downloaded and/or printed for private study, or other acts as permitted by national copyright laws. The publisher or other rights holders may allow further reproduction and re-use of the full text version. This is indicated by the licence information on the White Rose Research Online record for the item.

Takedown

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing eprints@whiterose.ac.uk including the URL of the record and the reason for the withdrawal request.

1 Fabrication of magnetic tunnel junctions with a metastable bcc 2 Co₃Mn disordered alloy as a bottom electrode

3 Kazuma KUNIMATSU^{1,2}, Tomoki TSUCHIYA^{3,4*}, Kelvin ELPHICK⁵ Tomohiro ICHINOSE²,
4 Kazuya Z. SUZUKI^{2,4}, Atsufumi. HIROHATA⁵, Shigemi MIZUKAMI^{2,3,4†},

5 ¹Department of Applied Physics, Graduate School of Engineering, Tohoku University, Sendai 980-8579,
6 Japan ²WPI Advanced Institute for Materials Research, Tohoku University, Katahira 2-1-1, Sendai
7 980-8577, Japan ³Center for Science and Innovation in Spintronics (CSIS), Core Research Cluster (CRC),
8 Tohoku University, Sendai 980-8577, Japan ⁴Center for Spintronics Research Network (CSRN), Tohoku
9 University, Sendai 980-8577, Japan ⁵Department of Electronic Engineering, The University of York, York
10 YO10 5DD, England

11
12 We fabricated MgO barrier magnetic tunnel junctions (MTJs) with a Co₃Mn alloy bottom and FeCoB
13 top electrodes. The (001)-oriented epitaxial films of the metastable bcc Co₃Mn disordered alloys obtained
14 showed saturation magnetization of approximately 1640 emu/cm³. The transmission electron microscopy
15 showed that the MgO barrier was epitaxially grown on the Co₃Mn electrode. Tunnel magnetoresistance of
16 approximately 150% was observed at room temperature after the annealing of MTJs at 350°C, indicating
17 that bcc Co₃Mn alloys have relatively high spin polarization.

18 The magnetic tunnel junction (MTJ) is a key device for spintronics,¹⁻³⁾ which has
19 been utilized in various magnetic sensors including the read head of a hard disk drive,
20 magnetoresistive random access memory, and neuromorphic applications.⁴⁻⁶⁾ One of the
21 issues is to enhance the tunnel magnetoresistance (TMR) effect, i.e., junction resistance
22 change depending on the parallel and antiparallel states of two magnetizations for the
23 junctions. Currently, the MgO barrier and FeCoB alloy electrodes are used as the stan-
24 dard MTJ barrier and magnetic materials,⁷⁻¹¹⁾ which exhibited the record 604% in the
25 TMR ratio at room temperature (RT).¹²⁾ Such a high TMR ratio is attributed to the
26 orbital symmetry filtering by the MgO barrier and the highly spin polarized Δ_1 band
27 in FeCo alloys.^{13,14)} To search for routes to further enhance the TMR ratio, it is curious
28 to investigate various magnetic metals other than FeCo binary systems.

29 Here, we report the TMR effect observed in MTJs utilizing different types of dis-

*E-mail: tomoki.tsuchiya.d1@tohoku.ac.jp

†E-mail: shigemi.mizukami.a7@tohoku.ac.jp

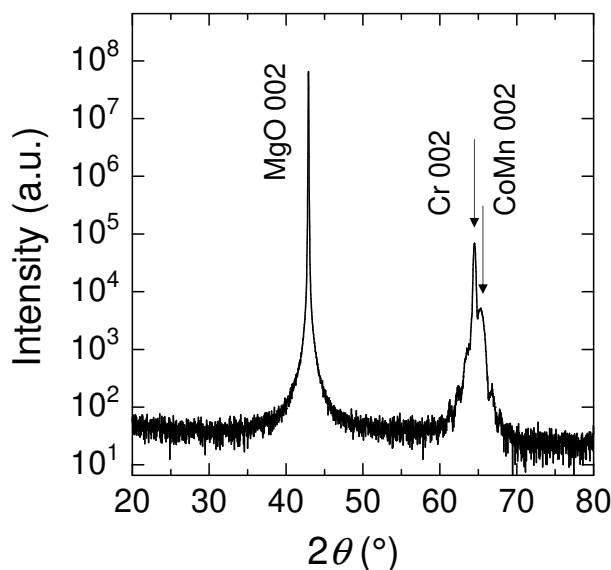


Fig. 1. Out-of-plane XRD pattern for Co_3Mn film deposited on (001) Cr-buffered MgO substrate.

1 ordered bcc CoMn alloy. A bulk Co-rich CoMn binary disordered alloy has a hcp or
 2 fcc phase as thermodynamically stable phase.^{15–18)} The saturation magnetizations and
 3 Curie temperatures decrease with increasing Mn concentration, and a magnetic long
 4 range order is lost around the Mn concentration of 30–35%.^{15–18)} In contrast, metastable
 5 bcc phase of Co-rich CoMn alloys show the relatively high saturation magnetization at
 6 similar Mn composition,^{19–21)} and a net magnetic moment per atom is in $2.32\text{--}2.53 \mu_B$
 7 at Mn concentration of 24%,²¹⁾ being close to that of a bcc Fe. This bcc phase is ob-
 8 tained in thin films grown on (001) GaAs and (001) MgO single crystalline substrates by
 9 molecular beam epitaxy (MBE) technique, as reported by a few groups.^{19–21)} However,
 10 there are no reports on MTJs comprised of bcc CoMn alloy electrodes to date.

11 All samples were deposited on (100) MgO single crystal substrates using a magnetron
 12 sputtering technique. The base pressure was 2×10^{-7} Pa. The MTJ staking structure was
 13 substrate/ Cr(40)/ Co_3Mn (10)/ Mg(0.4)/ MgO(2)/ $\text{Fe}_{60}\text{Co}_{20}\text{B}_{20}$ (4.5)/ Ta(3)/ Ru(5)
 14 (thickness in nm). All layers were deposited at RT. The composition of Co_3Mn film
 15 is $\text{Co}_{74}\text{Mn}_{26}$ (at.%) determined using inductively-coupled plasma mass spectrometer.
 16 We also prepared samples of substrate/ Cr(40)/ Co_3Mn (10)/ Mg(0.4)/ MgO(2)/ Ta(2)
 17 for structural and magnetization measurements. The crystal structures of the samples
 18 were determined using an x-ray diffractometer (XRD) by Cu K_α radiation. Nanostruc-
 19 tural analysis of samples was conducted by transmission electron microscopy (TEM).
 20 Magnetization measurements were performed using a vibrating sample magnetometer.

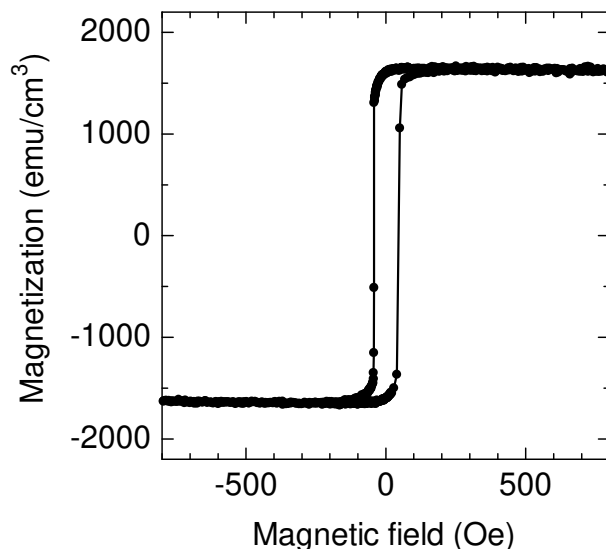


Fig. 2. In-plane magnetization hysteresis loop for the Co_3Mn film deposited on (001) Cr buffered MgO substrate.

1 The microfabrication of the MTJs were performed using a standard ultraviolet photo-
 2 lithography and Ar ion milling. The thirty six junctions with rectangular shapes were
 3 obtained on the substrate with the junction areas of 60×15 , 40×10 , 20×5 , 40×2 , 15×3 ,
 4 and $20 \times 2 \mu\text{m}^2$. The MTJs were annealed with a vacuum furnace at the temperature
 5 range $250\text{--}400^\circ\text{C}$. Magnetoresistance (MR) for the MTJs was measured by a four-probe
 6 method using a prober system with a maximum applied field of approximately 1 kOe.
 7 All the measurements were performed at RT.

8 Out-of-plane XRD pattern of the Co_3Mn film is shown in Fig. 1. The 002 peaks from
 9 the Cr buffer layer and bcc Co_3Mn were observed, but no other peaks, in particular
 10 those from fcc Co-Mn, were detected. The out-of-plane lattice parameter for the Co_3Mn
 11 film was evaluated as approximately 0.286 nm, which is close to the lattice constant
 12 for the bcc Co_3Mn of 0.285 nm.¹⁹⁾ Thus, it is considered that the (001)-oriented bcc
 13 Co_3Mn films were obtained on (001) Cr-buffered MgO substrates.

14 The in-plane magnetization curve is shown in Fig. 2. The saturation magnetization
 15 M_s is approximately 1640 emu/cm^3 . This value is comparable to that of Co or Fe
 16 and is also similar to the magnetic moment value evaluated by x-ray magnetic circular
 17 dichroism for bcc $\text{Co}_{76}\text{Mn}_{24}$ alloy films,²¹⁾ rather than that of fcc CoMn alloys with the
 18 similar Mn concentration.¹⁶⁾

19 The MR curves measured at RT for the $40 \times 2 \mu\text{m}^2$ MTJ annealed at 350°C is shown
 20 in Fig. 3(a). The resistance changes depending on the magnetization configuration are

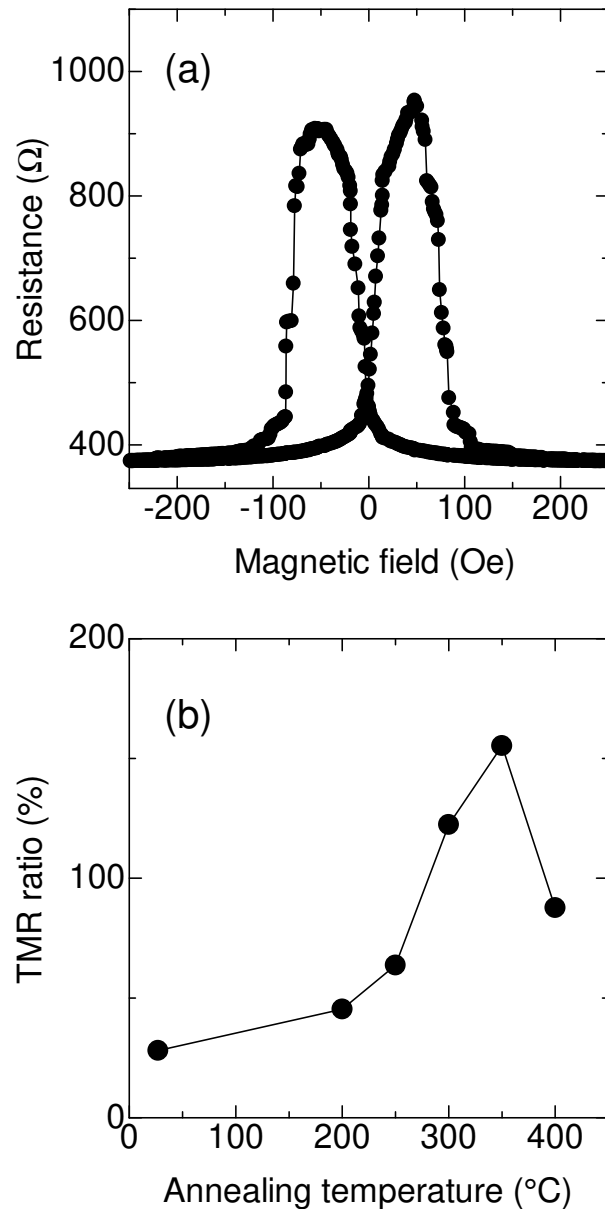


Fig. 3. (a) The typical MR curve for MTJs with Co_3Mn film as the bottom electrode. (b) The TMR ratio as a function of the annealing temperature of the MTJ with Co_3Mn film as the bottom electrode.

1 observed. Note that the MTJ is a pseudo-spin valve type, which means that both
2 magnetic layers were unpinned by the exchange bias¹²⁾ and the antiparallel state would
3 not be well defined in this study. Figure 3(b) shows the TMR ratio for this junction
4 as a function of the annealing temperature of the MTJ. The maximum TMR ratio was
5 observed as 155% at the annealing temperature of 350 $^\circ\text{C}$ in Fig. 3(b) and was 158%
6 for the different MTJ on the same substrate. This value is smaller than the TMR ratio
7 of $\sim 200\%$ observed at RT in Fe/MgO/Fe fully-epitaxial MTJs fabricated by the MBE

1 technique.²²⁾

2 Figure 4 shows the cross sectional TEM image for the MTJ sample annealed at
3 350°C. The MgO barrier is epitaxially grown on the bcc (001) Co₃Mn electrode. More-
4 over, the coherency of the lattices of Co₃Mn, MgO, and almost crystallized FeCoB at
5 the bottom and top interfaces are visible. These observations mean that the coherent
6 tunneling is expected if the bcc Co₃Mn has the Δ_1 band at the Fermi level.

7 To gain insight into the spin polarization of the bcc Co₃Mn studied here, Julliere's
8 model was used for approximate estimation, which can be expressed as¹⁾

$$\text{TMR ratio (\%)} = \frac{2P_1P_2}{1 - P_1P_2} \times 100, \quad (1)$$

9 where P_1 and P_2 are the tunneling spin polarization for each magnetic electrode. Since
10 this relation is hold only for an incoherent tunneling, the evaluate spin polarization
11 should be regarded as an effective value in case of the coherent tunneling. To account
12 for the TMR ratio observed in this study using this relation, the tunneling spin polar-
13 ization for bcc Co₃Mn with MgO barrier should be **at least 0.44 at RT if the tunneling**
14 **spin polarization of FeCoB is 1** . This is relatively higher than the spin polarization
15 of 0.33 evaluated at low temperature in Co₇₃Mn₂₇ alloy, which had a low saturation
16 magnetization and was unlikely bcc phase.²³⁾ A more detailed discussion is beyond the
17 scope of this brief report and will be provided elsewhere.

18 In summary, we fabricated Co₃Mn/MgO/FeCoB MTJs using the sputtering tech-
19 nique. The (001)-oriented metastable bcc Co₃Mn epitaxial films obtained exhibited
20 saturation magnetization of approximately 1640 emu/cm³. The cross-sectional TEM
21 showed that the MgO barrier was epitaxially grown on the Co₃Mn electrode. We ob-
22 served the TMR ratio of 158% at RT for MTJs annealed at 350°C, indicating that
23 metastable bcc Co₃Mn alloys have relatively high spin polarization.

24 **Acknowledgments** We would like to thank Y. Kondo for his assistance. This work was partially
25 supported by JST CREST (No. JPMJCR17J5).

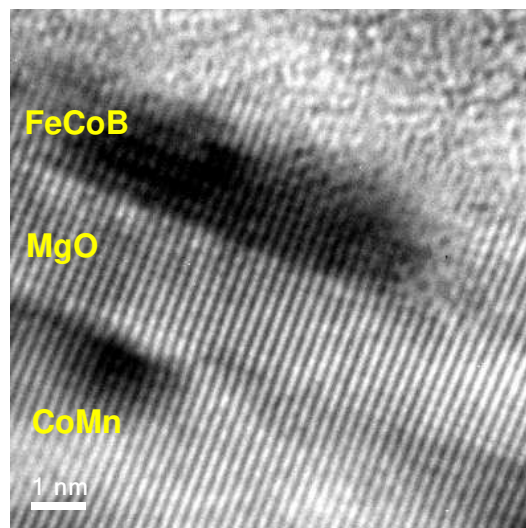


Fig. 4. The cross-sectional TEM image for the MTJ sample.

1 **References**

- 2 1) M. Julliere, Phys. Lett. A **54**, 225 (1975).
- 3 2) T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).
- 4 3) J.S. Moodera, L.R. Kinder, T.M. Wong, and R. Meservey, Phys. Rev. Lett. **74**,
- 5 3273 (1995).
- 6 4) J.-G. (Jimmy) Zhu and C. Park, Mater. Today **9**, 36 (2006).
- 7 5) A.D. Kent and D.C. Worledge, Nat. Nanotechnol. **10**, 187 (2015).
- 8 6) M. Romera, P. Talatchian, S. Tsunegi, F. Abreu Araujo, V. Cros, P. Bortolotti, J.
- 9 Trastoy, K. Yakushiji, A. Fukushima, H. Kubota, S. Yuasa, M. Ernoult, D.
- 10 Vodenicarevic, T. Hirtzlin, N. Locatelli, D. Querlioz, and J. Grollier, Nature **563**,
- 11 230 (2018).
- 12 7) S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nat. Mater. **3**,
- 13 868 (2004).
- 14 8) S.S.P. Parkin, C. Kaiser, A. Panchula, P.M. Rice, B. Hughes, M. Samant, and
- 15 S.-H. Yang, Nat. Mater. **3**, 862 (2004).
- 16 9) S. Yuasa, T. Katayama, T. Nagahama, A. Fukushima, H. Kubota, Y. Suzuki, and
- 17 K. Ando, Appl. Phys. Lett. **87**, 222508 (2005).
- 18 10) S. Yuasa, A. Fukushima, H. Kubota, Y. Suzuki, and K. Ando, Appl. Phys. Lett.
- 19 **89**, 042505 (2006).
- 20 11) D.D. Djayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yamagata, N.
- 21 Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **86**, 092502
- 22 (2005).
- 23 12) S. Ikeda, J. Hayakawa, Y. Ashizawa, Y.M. Lee, K. Miura, H. Hasegawa, M.
- 24 Tsunoda, F. Matsukura, and H. Ohno, Appl. Phys. Lett. **93**, 082508 (2008).
- 25 13) J. Mathon and A. Umerski, Phys. Rev. B **63**, 220403 (2001).
- 26 14) W.H. Butler, X.-G. Zhang, T.C. Schulthess, and J.M. MacLaren, Phys. Rev. B
- 27 **63**, 054416 (2001).
- 28 15) J.S. Kouvel, J. Phys. Chem. Solids **16**, 107 (1960).
- 29 16) M. Matsui, T. Ido, K. Sato, and K. Adachi, J. Phys. Soc. Japan **28**, 791 (1970).
- 30 17) A. Z. Menshikov, G. A. Takzei, Yu. A. Dorofeev, V. A. Kazan- stev, A. K.
- 31 Kostyshin, and I. I. Sych, Zh. Eksp. Teor. Fiz. **89**, 1269 (1985) [Sov. Phys. JETP
- 32 **62**, 734 (1985)].
- 33 18) K. Ishida and T. Nishizawa, Bull. Alloy Phase Diagrams **11**, 125 (1990).

- 1 19) D. Wu, G.L. Liu, C. Jing, Y.Z. Wu, D. Loison, G.S. Dong, X.F. Jin, and D.-S.
2 Wang, Phys. Rev. B **63**, 214403 (2001).
- 3 20) L. Zhang, D. Basiaga, J.R. O'Brien, and D. Heiman, J. Appl. Phys. **98**, 106101
4 (2005).
- 5 21) R.J. Snow, H. Bhatkar, A.T. N'Diaye, E. Arenholz, and Y.U. Idzerda, J. Magn.
6 Magn. Mater. **419**, 490 (2016).
- 7 22) F. Bonell, T. Hauet, S. Andrieu, F. Bertran, P. Le Fèvre, L. Calmels, A. Tejada,
8 F. Montaigne, B. Warot-Fonrose, B. Belhadji, A. Nicolaou, and A.
9 Taleb-Ibrahimi, Phys. Rev. Lett. **108**, 176602 (2012).
- 10 23) T.H. Kim and J.S. Moodera, Phys. Rev. B **66**, 104436 (2002).