Extended Abstracts of the 2007 International Conference on Solid State Devices and Materials, Tsukuba, 2007, pp. 744-745

C-5-2 (Invited)

Structural Study on CoFeB/MgO/CoFeB Magnetic Tunnel Junctions

K. Tsunekawa,[1,2] Y. Choi,[1] Y. Nagamine,[1] H. Maehara,[1]
D. D. Djayaprawira,[1] T. Takeuchi,[2] and Y. Kitamoto[2]

[1] Electronic Devices Engineering Headquarters, Canon ANELVA Corporation
5-8-1, Yotsuya, Fuchu-shi, Tokyo 183-8508, Japan
Phone: +81-42-334-0269 E-mail: tsunekawa.koji@canon-anelva.co.jp
[2] Dept. of Innovative and Engineered Materials, Tokyo Institute of Technology
4259 Nagatsuta-cho, Midori-ku, Yokohama 226-8502, Japan

1. Introduction

Crystalline MgO-based magnetic tunnel junctions (MTJs) have attracted much attention due to their large room-temperature magnetoresistance (MR) over 100% [1-4] and potential applications to magnetic random access memories [5-6] and read sensors in high-density magnetic recording [7-8]. Giant tunneling magnetoresistance (TMR) of several 1000% in MgO-based MTJs was theoretically predicted in the fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJ in 2001 [9-10], and TMR of 180% at room temperature (RT) was experimentally demonstrated in a fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJ fabricated by molecular beam epitaxy [1]. Subsequently, Djayaprawira *et al.* reported that non-epitaxial CoFeB/MgO/CoFeB MTJs prepared by magnetron sputtering method also shows a very large MR ratio more than 200% at RT [3]. The impact of the report on practical application is that the MgO-based MTJ using CoFeB was prepared onto a thermally oxidized Si wafer by the magnetron sputtering method, which enlightened the prompt applicability to the mass production.

To date, three possible explanations have been suggested on the mechanism of the large TMR on the CoFeB/MgO/CoFeB MTJs [3-4, 11-12]: i) CoFeB containing about 20 at.% boron is amorphous in the as-deposited state, ii) MgO grows with (001) texture on amorphous CoFeB layer, and iii) the amorphous CoFeB layers apparently crystallize after high-temperature annealing and their crystalline orientation is bcc (001) out-of-plane. Based on these microstructure studies, it can be considered that the crystalline structure of bcc CoFe(001)/MgO(001)/bcc CoFe(001) is responsible for high TMR as predicted in the epitaxial Fe(001)/MgO(001)/Fe(001) system. In order to satisfy this, besides the out-of-plane (001) texture, the in-plane rotational epitaxial relationship also is required. Here, we report our detailed study on the crystallization process of CoFeB ferromagnetic layers above and below the MgO barrier layer in MTJs fabricated by the magnetron sputtering and clarified the epitaxial relationship between crystallized CoFe and MgO.

2. Experimental

An MTJ sample with a stack configuration of Ta(10 nm)/$Pt_{49}Mn_{51}$(15 nm)/$Co_{70}Fe_{30}$(2.5 nm)/Ru(0.85 nm)/$(Co_{75}Fe_{25})_{80}B_{20}$(3 nm)/Mg(0.3 nm)/MgO(1.5 nm)/$(Co_{75}Fe_{25})_{80}B_{20}$(3 nm)/Ta(10 nm)/Ru(7 nm) was deposited onto thermally oxidized Si(001) wafer using a magnetron sputtering system (Canon ANELVA C-7100). The base pressure before deposition was lower than 7×10^{-7} Pa. All the metallic layers in the stacks were deposited by dc sputtering in Ar or Kr atmosphere, and the MgO layer was deposited directly from a sintered MgO target by rf sputtering in an Ar atmosphere. The substrate temperature during deposition was maintained at RT by indirect water-cooling. After deposition, the sample was vacuum-annealed at 360°C for 2 h in a magnetic field of 8 kOe in order to induce a chemical ordering of the antiferromagnetic PtMn layer and a crystallization of the amorphous CoFeB layers. The MR ratio and RA (resistance area product) were characterized by current-in-plane tunneling (CIPT) method [13]. Structural analysis of the sample was carried out by high-resolution transmission electron microscopy (HRTEM).

3. Results

Figure 1(a) shows a cross-section HRTEM image of the MgO-based MTJ whose MR and RA were found to be 213% and 11359 $\Omega\mu m^2$, respectively, by the CIPT method. It can be confirmed that the MgO layer is highly textured polycrystalline with (001) out-of-plane preferred orientation, and the interfaces between layers are atomically sharp and flat. Based on the atomic plane stacking sequences, either AAA… or ABABAB…, and measuring the interatomic spacing, it is possible to define the in-plane orientation of MgO grains shown in squared area (b) and (c), which are pointing out of panel along with [100] and [110], respectively. The in-plane crystallographic directions of CoFeB layer in the squared area (b) and (c) are confirmed to be [110] and [100], respectively, by atomic stacking sequences and interatomic spacing measurement. It is further verified that this in-plane crystallographic direction is closely related to that of MgO layer, and furthermore grain boundary of crystallized CoFeB coincides with that of MgO layer. Considering crystallography of these layers, we suggest the grain-to-grain epitaxy through a 45° rotational epitaxy rather than cube-on-cube epitaxy between MgO grain and crystallized CoFe grain, in that bcc CoFe(001)[110]// MgO(001)[100]. The rotational epitaxy is the only way to reduce a lattice mismatch down to 4.9% from a huge lattice mismatch of about 32% between MgO (a = 0.421 nm) and $Co_{75}Fe_{25}$ (a = 0.283 nm). Figures 1(d) and 1(e) show the corresponding schematic projections to the lattice patterns seen in Figs. 1(b) and 1(c), respectively. Each of these schematics represents the pseudomorphic epitaxial grains when the electron beam is projected parallel to CoFe[110]//MgO[100] and CoFe[010]//MgO[110]. This means that the grain-to-grain epitaxy is realized between the crystallized bcc CoFe and the (001) textured MgO layers by the 45° rotational epitaxial relationship and a large number of pseudomorphic epitaxial columnar grains with the grain-to-grain epitaxy exist in the CoFeB/MgO/CoFeB MTJ. In short, the epitaxial relationship of bcc CoFe(001)[110]//MgO(001)[100] is realized within each columnar grain, which is satisfying the structural prerequisite of coherent tunneling predicted in the theories [9-10]. The detail of the 45° rotational epitaxy between the crystallized bcc CoFe and the (001)-textured MgO layers is explained elsewhere [14].

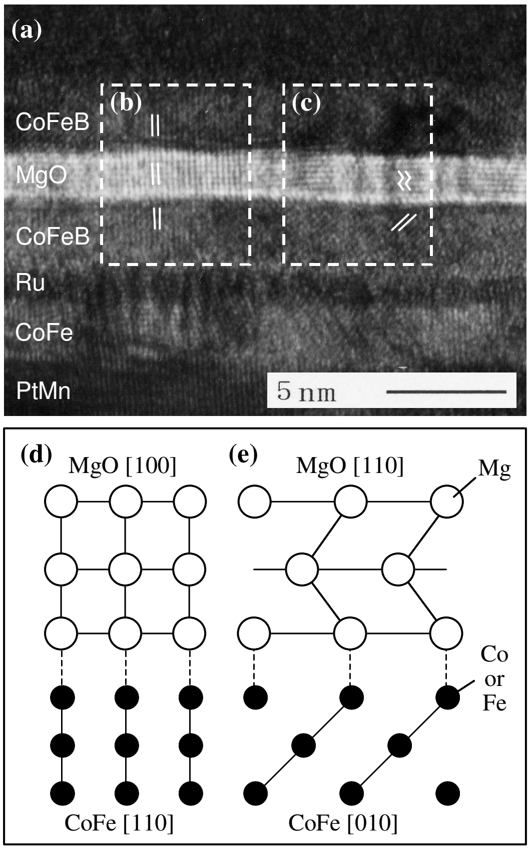

Fig. 1 (a) Cross-section HRTEM image of the CoFeB/MgO/CoFeB MTJ annealed at 360°C for 2 h.. Diagrams (d) and (e) show the corresponding schematic projections of the lattice patterns seen in areas (b) and (c) in the HRTEM image, respectively. The indices represent the azimuthal direction for each crystal.

3. Conclusions

We showed the existence of the pseudomorphic epitaxial columnar grains in the CoFeB/MgO/CoFeB MTJ, each of which has an epitaxial relation ship of bcc CoFe(001)[110]//MgO(001)[100]//bcc CoFe(001)[110]. It originates that CoFe crystallizes out from the interface with the (001) textured MgO layer by solid-state nucleation and grain growth during high-temperature annealing. Within each of this columnar grain, we suggest that the coherent tunneling, which is a prerequisite for huge TMR predicted by theoretical calculation, is realized.

Acknowledgements

We are grateful to all the members of the C-7100 process team at Canon ANELVA for their effort in preparing and characterizing samples.

References

[1] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nature Mater. **3** (2004) 868.

[2] S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang, Nature Mater. **3** (2004) 862.

[3] D. D. Djayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yamagata, N. Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **86** (2005) 092502.

[4] J. Hayakawa, S. Ikeda, F. Matsukura, H. Takahashi, and H. Ohno, Jpn. J. Appl. Phys. **44** (2005) L587.

[5] S. Tehrani, J. M. Slaughter, E. Chen, M. Durlam, J. Shi, and M. DeHerrera, IEEE Trans. Magn. **35** (1999) 2814.

[6] S. S. P. Parkin, K. P. Roche, M. G. Samant, P. M. Rice, R. B. Beyers, R. E. Scheuerlein, E. J. O'Sullivan, S. L. Brown, J. Bucchigano, D. W. Abraham, Yu Lu, M. Rooks, P. L. Trouilloud, R. A. Wanner, and W. J. Gallagher, J. Appl. Phys. **85** (1999) 5828.

[7] S. Araki, K. Sato, T. Kagami, S. Saruki, T. Uesugi, N. Kasahara, T. Kuwashima, N. Ohta, J. Sun, K. Nagai, S. Li, N. Hachisuka, H. Hatate, T. Kagotani, N. Takahashi, K. Ueda, and M. Matsuzaki, IEEE Trans. Magn. **38** (2002) 72.

[8] S. Mao, Solid State Technology, September (2004) S11.

[9] W. H. Butler, X.-G. Zhang, T. C. Schulthess and J. M. MacLaren, Phys. Rev. B **63** (2001) 054416.

[10] J. Mathon and A. Umerski, Phys. Rev. B **63** (2001) 220403R.

[11] S. Yuasa, Y. Suzuki, T. Katayama and K. Ando, Appl. Phys. Lett. **87** (2005) 252503.

[12] C. Park, J. G. Zhu, M. T. Moneck, Y. Peng and D. E. Laughlin, J. Appl. Phys. **99** (2006) 08A901.

[13] C. Worledge and P. L. Trouilloud, Appl. Phys. Lett. **83** (2003) 84.

[14] Y. Choi, K. Tsunekawa, Y. Nagamine, and D. D. Djayaprawira, J. Appl. Phys. **101** (2007) 013907.