Magnetic tunnel junction with a metastable bcc Co₃Mn alloy electrode

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Abstract

We fabricated bcc-Co₃Mn/MgO/CoFeB magnetic tunnel junctions (MTJs). The (001)-oriented epitaxial films of the metastable bcc Co₃Mn disordered alloys were successfully prepared on MgO(100) substrates by a sputtering technique. It showed saturation magnetization of approximately 1640 emu/cm³, indicating the ferromagnetic coupling between Co and Mn. The transmission electron microscopy showed that the MgO barrier was epitaxially grown on the Co₃Mn electrode. Tunnel magnetoresistance (TMR) of > 150% was observed at room temperature (RT). This is a first observation of the relatively large TMR effect at RT without using conventional FeCo based alloys or Heusler alloys.

1. Introduction

Magnetic tunnel junction (MTJ) is one of the key spintronics devices. MTJs have a crucial role for various magnetic sensors, a hard disk drives, magnetoresistive random access memory (MRAM), and future neuromorphic applications. Although there are several issues for developing advanced MTJs depending on the applications mentioned above, one common issue is an enhancement of the tunnel magnetoresistance (TMR) effect, i.e., junction resistance change depending on the parallel and antiparallel states of two magnetizations for the junctions. Currently, the MgO barrier and CoFe(B) alloy electrodes are used as the standard MTJ barrier and magnetic materials, respectively [1], which exhibited the record 604% in the TMR ratio at room temperature (RT) [2]. To further enhance the TMR ratio, it is of importance to search different magnetic metals other than CoFe-based alloys. The high TMR ratio observed in CoFe(B)/MgO system is attributed to the orbital symmetry filtering by the MgO barrier and the fully spin polarized Δ_1 band in Fe or Fe rich FeCo alloys. Another candidate is bcc Co because bcc Co also have the fully spin polarized Δ_1 band and showed a large TMR ratio [3]. However, bcc Co can be obtained only as a few monolayer thick ultrathin film with a forced epitaxy.

Bcc Co-Mn alloys are interesting candidates. A thermodynamically stable phase for Co-Mn binary alloy is a disordered hcp or fcc structure. The saturation magnetizations and Curie temperatures are reduced as Mn concentration increases. A magnetic long-range order is lost around the Mn concentration of 30–35% [4]. On other hand, bcc CoMn alloy can be obtained as a thin film when those are grown on GaAs(001) and MgO(001) single crystalline substrates with molecular beam epitaxy (MBE) technique, though only a few groups reported [5,6]. However, there are no reports on film growth with a sputtering technique as well as MTJ fabrication comprising bcc CoMn alloy electrodes.

2. Experimental procedures

All samples were prepared using (100) MgO single crystal substrates and the magnetron sputtering. The base pressure was 2×10^{-7} Pa. The MTJ staking structure was Cr(40)/ Co₃Mn(10)/ Mg(0.4)/ MgO(2)/ CoFeB(4.5)/ Ta(3)/ Ru(5) (thickness in nm). The composition of Co₃Mn film was determined as Co₇₄Mn₂₆ (at.%) using an inductively coupled plasma mass spectrometer. The samples of only the bottom electrode films were also fabricated to characterize crystal structure and magnetic properties. The crystal structures of the samples were determined using an x-ray diffractometer (XRD) by Cu K_{α} radiation and the transmission electron microscopy (TEM). Magnetization measurements were performed using a vibrating sample magnetometer (VSM). The micro-fabrications of the MTJs were performed using a standard ultraviolet photo-lithography and Ar ion milling. The 36 junctions with rectangular shapes were obtained on the substrate with the junction areas of 60×15 , 40×10 , 20×5 , 40×2, 15×3, and 20×2 μ m². The MTJs were annealed with a vacuum furnace at the temperature range 250-400°C. Magnetoresistance (MR) for the MTJs was measured by a four-probe method. All the measurements were performed at RT.

3. Results and discussion

Out-of-plane XRD pattern of the Co_3Mn film is shown in Fig 1. The 002 peaks from the Cr buffer layer and bcc Co_3Mn layer were observed. On the other hand, the diffraction peaks from fcc CoMn were not detected. The out-of-plane lattice parameter for the Co_3Mn film was evaluated as approximately 0.286 nm, which is close to the lattice constant reported for the bcc Co_3Mn of 0.285 nm [5]. Thus, it is considered that the (001)-oriented bcc Co_3Mn films were obtained on (001) Cr-buffered MgO substrates.

Past x-ray magnetic circular dichroism studies reported the net magnetic moment for the bcc CoMn alloy films was 2.32–2.53 μ_B /atom at the Mn concentration of 24%. This magnetic moment value is close to that of a bcc Fe [6]. The VSM measurements for our bcc Co₃Mn film showed a well-defined magnetization hysteresis loop and the saturation magnetization M_s of approximately 1640 emu/cm³. This magnetization value is comparable to the magnetic moment mentioned above and is considerably larger than that of fcc CoMn alloys with the similar Mn concentration [6].

The representative TMR curve measured at RT for the $20 \times 2 \ \mu m^2$ MTJ annealed at 350°C is shown in Fig. 2. The resistance changes are observed depending on the magnetization configuration. The antiparallel magnetization configuration is not well defined because the both magnetic layers in the MTJs were unpinned by the exchange bias. The maximum TMR ratio was observed as 158% at the annealing temperature of 350°C. According to the cross sectional TEM image for the MTJ sample annealed at 350°C, the MgO barrier was epitaxially grown on the (001) surface of the bcc Co₃Mn electrode. Moreover, the coherency of the lattices of Co₃Mn, MgO, and almost crystallized CoFeB were clearly identified. These observations mean that the coherent tunneling is expected if the bcc Co₃Mn disordered alloys has the Δ_1 band at the Fermi level.

It may be valuable to comment on the spin polarization of the bcc Co₃Mn studied here. Julliere's model [7] was used for approximate estimation of the spin polarization even though the coherent tunneling regime and it is expressed as,

TMR ratio (%) =
$$\frac{2P_{COMn}P_{CoFeB}}{1-P_{COMn}P_{CoFeB}} \times 100$$

Here P_{CoMn} and P_{CoFeB} are the tunneling spin polarization for CoMn and CoFeB electrode, respectively. From this relation, the lower bound of the tunneling spin polarization for bcc Co₃Mn with MgO barrier was obtained as 0.44 at RT with setting P_{CoFeB} of 1. True value of P_{CoMn} would be much higher than 0.44, which is distinct from the spin polarization of 0.33 for fcc Co₇₃Mn₂₇ alloy [8].

4. Conclusions

Co₃Mn/MgO/CoFeB MTJs were fabricated using the sputtering technique. The (001)-oriented metastable bcc Co₃Mn epitaxial films exhibited the saturation magnetization of approximately 1640 emu/cm³. The cross-sectional TEM showed that the MgO barrier was epitaxially grown on the Co₃Mn electrode. The TMR ratio of 158% at RT was observed for MTJs annealed at 350°C, indicating that metastable bcc CoMn alloys potentially have high spin polarization [9].

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References

- D.D. Djayaprawira et al., Appl. Phys. Lett. 86, 092502 (2005).
- [2] S. Ikeda et al., Appl. Phys. Lett. 93, 082508 (2008).
- [3] S. Yuasa et al., Appl. Phys. Lett **89**, 042505 (2006)
- [4] M. Matsui et al., J. Phys. Soc. Japan 28, 791 (1970).
- [5] D. Wu et al, Phys. Rev. B 63, 214403 (2001).
- [6] R.J. Snow et al., J. Magn. Magn. Mater. 419, 490 (2016).
- [7] M. Julliere, Phys. Lett. A 54, 225 (1975).
- [8] T.H. Kim and J.S. Moodera, Phys. Rev. B 66, 104436 (2002).
- [9] K. Kunimatsu and T. Tsuchiya et al., submitted (2019).



Figure 1: The out of plane X-ray diffraction pattern for the 10-nm-thick Co₃Mn film deposited on the Cr-buffered MgO substrate.



Figure 2: The tunnel magnetoresistance (TMR) curve measured at room temperature for the $Co_3Mn/MgO/CoFeB$ junction.