

Magnetic tunnel junctions with an equiatomic CoFeMnSi electrode

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Abstract

Equiatomic quaternary CoFeMnSi Heusler alloy is one of the candidates of spin-gapless materials. Here we studied tunnel magnetoresistance (TMR) in magnetic tunnel junctions (MTJs) comprising CoFeMnSi and CoFe alloy electrodes. The MTJs multilayer stacking were fabricated with ultrahigh-vacuum magnetron sputtering. Maximum TMR ratios of 101% and 521% for the MTJs were obtained at room and low temperatures, respectively. The large bias voltage dependence of the TMR ratio was also observed at low temperature. Spin polarization for CoFeMnSi was approximately evaluated to be 0.85 from the TMR ratio obtained at low temperature.

1. Introduction

Development of high spin polarization materials is one of the issues for spintronics, which are expected to be used for non-volatile, reconfigurable logic functions and ultralow power consumption. Half-metallic ferromagnets feature complete spin polarization P at the Fermi level E_F and are suitable candidates for spin-polarized current sources. Among the various half-metallic materials, Co_2YZ and quaternary $\text{Co}_2\text{Y}_{1-x}\text{Y}'_x\text{Z}$ full Heusler alloys have a special place due to their tunable electronic structure, a Curie temperature T_C that is well above room temperature (RT), and P that is close to unity [1-3].

As a different class of materials, there is increasing interest in equiatomic quaternary $\text{XX}'\text{YZ}$ Heusler alloys since these materials exhibit various properties resulting from their crystal structures differing in symmetry from the original Heusler $L2_1$ structure [4]. We focus here on an equiatomic quaternary CoFeMnSi alloy from among the numerous elemental combinations suggested [5]. Interestingly, CoFeMnSi alloy is one of the candidates of spin-gapless materials which have a half-metal band gap for a minority spin channel and a pseudo-gap for a majority spin channel [6], thus it can be regarded as a *half-metallic semimetal*. Quite recently, we obtained the single-crystalline CoFeMnSi films grown by sputtering method [7]. Therefore we study tunnel magnetoresistance (TMR) for magnetic tunnel junctions (MTJs) using the single crystalline CoFeMnSi electrode and discuss its spin polarization [8].

2. Experimental details

All the samples were prepared using an ultrahigh-vacuum magnetron sputtering system with a base pressure below

2×10^{-7} Pa. The MTJs multilayer stacking structure was (001) single crystalline MgO substrate/ Cr(40)/ CoFeMnSi(30)/ Mg(0.4)/ MgO(1.6 or 2.0)/ CoFe(5)/ IrMn(10)/Ta(3)/Ru(5) (thickness is in nm). The composition of the CoFeMnSi films was confirmed to be $\text{Co}_{25.3}\text{Fe}_{24.3}\text{Mn}_{24.0}\text{Si}_{26.4}$ (at.%) using inductively coupled plasma mass spectrometry. All the layers were deposited at RT. The CoFeMnSi layers were prepared using *in-situ* annealing at temperatures T_a^{CFMS} of 773, 873, and 973 K. The details of the fabrication of CoFeMnSi films are described elsewhere [7,8]. The microfabrication of MTJs with junction areas ranging from 10×10 to $100 \times 100 \mu\text{m}^2$ was performed using the standard ultraviolet photo-lithography and Ar ion milling. Following the microfabrication, *ex-situ* annealing was performed in the temperature range T_a^{MTJ} of 523-773 K under an in-plane magnetic field of 5 kOe. Transport properties were investigated using a four-probe method using a prober system with a maximum applied field of 2 kOe at RT and using a physical property measurement system (PPMS, Quantum Design) in the temperature range T of 10-300 K with an applied magnetic field of up to 3 kOe. The bias voltage V dependence of the TMR effect were measured using a current source (Keithley, 6221) and nano-voltmeter (Keithley, 2182A) [8].

3. Results and discussion

We investigated the influences of annealing CoFeMnSi electrode and MTJs on the TMR effect in MTJs with a nominal MgO barrier thickness of 2.0 nm, *i.e.*, Mg(0.4)/MgO(1.6). We obtained a well-defined anti-parallel configuration of magnetization at $T_a^{\text{MTJ}} > 573$ K. The TMR ratio increased with increasing T_a^{MTJ} , and it showed the maximum value at $T_a^{\text{MTJ}} = 723$ K. The maximum TMR ratio of 89% was obtained in the MTJ for $T_a^{\text{CFMS}} = 873$ K, which was consistent with the fact that the CoFeMnSi electrode annealed at $T_a^{\text{CFMS}} = 773-873$ K showed the highest $B2$ and $L2_1$ chemical ordering and atomically flat surfaces. The increase of the TMR ratio with increasing T_a^{MTJ} was similarly observed in various MgO-based MTJs [9,10], mostly due to the improvement in the crystallinity and texture of the MgO barrier and in the interface structures [8].

Figures 1(a) and 1(b) show the TMR curves and the temperature dependence of the TMR ratio for the MTJ with the 2.4-nm-thick MgO barrier. The TMR ratio at $T = 300$ K is 101%, and it remarkably increases with decreasing temperature, up

to 521% at $T = 10$ K, as shown in Fig. 1(b). We also investigated the temperature dependence of the TMR ratio for the MTJs with the 2.0-nm-thick MgO barrier. These MTJs also showed large increases in the TMR ratio at low temperature, similar to the data shown in Fig. 1. The TMR ratios for those MTJs with $T_a^{\text{CFMS}} = 873$ (973) K were 405 (345) % at $T = 10$ K, which was slightly smaller than that in Fig. 1. The TMR ratio rapidly decreased with increasing the absolute value of the bias voltage V in the regime of $V = \pm 0.1$ V [8]. This large bias voltage dependence of the TMR ratio was similarly observed in Co_2MnSi Heusler alloys-based MTJs in the past [2].

In order to gain insight into the half-metallicity for CoFeMnSi in this study, we try to evaluate spin polarization for CoFeMnSi from the TMR ratio at low temperature in the present study. In our MTJs the symmetry filtering and coherent tunneling effect is less pronounced [8], so that the Julliere's model is approximately used for the evaluation [11]. The TMR ratio can be expressed as $\text{TMR ratio (\%)} = 2P_{\text{CoFeMnSi}}P_{\text{CoFe}}/(1 - P_{\text{CoFeMnSi}}P_{\text{CoFe}}) \times 100$, where P_{CoFeMnSi} and P_{CoFe} are spin polarization for CoFeMnSi and CoFe, respectively, according to the model [11]. The value for P_{CoFe} is 0.5 for non-coherent tunneling, such as amorphous Al-O barrier-based MTJs [12], and then it were enhanced to 0.74-0.85 by symmetry filtering effect of MgO in coherent tunneling [9], leading to very high TMR effect.. If we assume P_{CoFe} of 0.85 in our MTJs, then the value for P_{CoFeMnSi} was evaluated to be approximately 0.85 from the TMR ratio observed at low temperature in this study. This P_{CoFeMnSi} value is comparable to the spin polarization for Co_2MnSi , 0.89 [1]. Therefore, it is considered that CoFeMnSi in this study has a high spin polarization and is close to a half-metal. The correlation between the pseudo-gap for the majority spin channel possibly present in this spin-gapless material and the spin-dependent transport for the MTJs will be a future subject [8].

4. Summary

We studied on the TMR effect for CoFeMnSi/MgO/CoFe MTJs to gain an insight into the half-metallicity of CoFeMnSi. A relatively high TMR ratio of about 521% and 101% were observed at 10 and 300 K, respectively. This suggested the high spin polarization of CoFeMnSi even though the obtained TMR ratio was smaller than the past record in the MgO-MTJs with CoFe and Mn-enriched Co_2MnSi Heusler alloy electrodes [13]. Spin polarization for CoFeMnSi was approximately evaluated to be 0.85 from the TMR ratio obtained at low temperature.

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References

- [1] Y. Sakuraba *et al.*, Jpn. J. Appl. Phys. **44** (2005) L1100.
- [2] Y. Sakuraba *et al.*, Appl. Phys. Lett. **89** (2006) 052508.

- [3] M. Yamamoto *et al.*, J. Phys. D: Appl. Phys. **39** (2006) 824.
- [4] L. Bainsla and K. G. Suresh, Appl. Phys. Rev. **3** (2016) 031101.
- [5] X. Dai *et al.*, J. Appl. Phys. **105** (2009) 07E901.
- [6] L. Bainsla *et al.*, Phys. Rev. B **91** (2015) 104408.
- [7] L. Bainsla *et al.*, Phys. Rev. B **96** (2017) 094404.
- [8] L. Bainsla *et al.*, Appl. Phys. Lett. **112** (2018) 052403.
- [9] S. S. P. Parkin *et al.*, Nat. Mater. **3** (2004) 862.
- [10] S. Tsunegi *et al.*, Appl. Phys. Lett. **93** (2008) 112506.
- [11] M. Julliere, Phys. Lett. **54A** (1975) 225.
- [12] D. J. Monsma *et al.*, Appl. Phys. Lett. **77** (2000) 720.
- [13] H. Liu *et al.*, Jpn. J. Appl. Phys. **51** (2012) 093004.

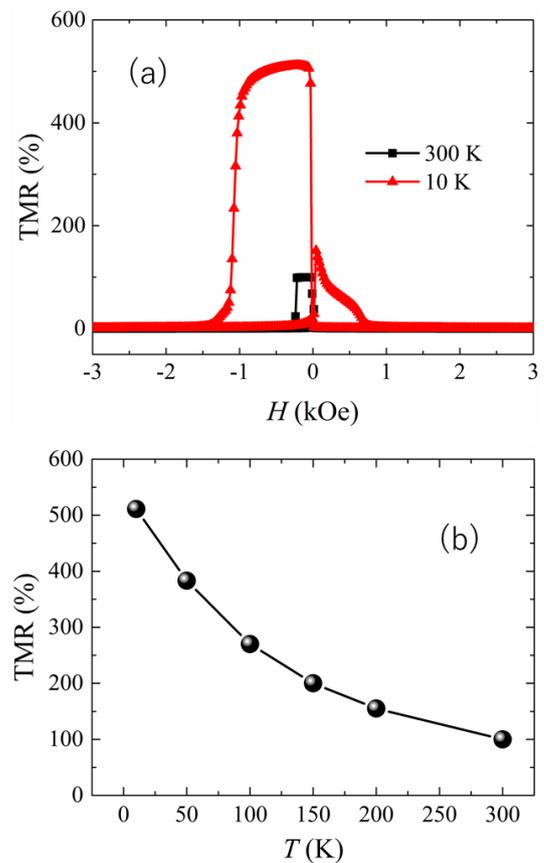


Fig. 1. (a) The TMR curves measured at room and low temperature for the CoFeMnSi/MgO/CoFe MTJs with $T_a^{\text{CFMS}} = 873$ K and $T_a^{\text{MTJ}} = 723$ K. (b) The TMR ratio as a function of temperature T . Here, T_a^{CFMS} and T_a^{MTJ} are the CoFeMnSi post annealing and MTJ *ex-situ* annealing temperatures, respectively.