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Magnetoresistance Characteristics of Magnetic Tunnel Junctions Consisting of Amorphous CoNbZr Alloys for Under and Capping Layers

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Magnetic tunnel junctions (MTJs) comprising amorphous CoNbZr layers have been investigated. $Co_{85.5}Nb_8Zr_{6.5}$ (in at. %) layers were employed to substitute the traditionally used Ta layers with an emphasis given on understanding underlayer effect. The typical junction structure was SiO_2/CoNbZr or Ta 2/CoFe 8/IrMn 7.5/CoFe 3/Al 1.6 + oxidation/CoFe 3/CoNbZr or Ta 2 (nm). For both as-deposited state and after annealing, the CoNbZr-underlayered structure showed superior surface smoothness up to the tunnel barrier than Ta-underlayerd one (rms roughness of 0.16 vs. 0.34 nm). CoNbZr-based MTJs was proven beneficial for increasing thermal stability and increasing V_h (the bias voltage where MR ratio becomes half) characteristics than Ta-based MTJs. This is because the CoNbZr-based junctions offer smoother interface structure than the Ta-based one.

Key words : Magnetic Tunnel Junction, Thermal Stability, Bias Voltage Dependence, CoNbZr, Surface Roughness

1. Introduction

Magnetic tunnel junctions (MTJs) have a large potential for use in a high areal density read head and magnetic random access memory (MRAM) applications because they exhibit large tunneling magnetoresistance (TMR) ratios [1, 2]. It has been observed that the junction resistance and TMR ratio are very sensitive to surface roughness of both the bottom electrode and the insulating layer [3]. In the MTJ, an insulating layer acts as a tunneling barrier of spin-polarized tunneling electrons between two ferromagnetic electrodes. The spin-dependent tunneling behavior is strongly affected not only by the spin-polarization of the both ferromagnetic electrodes but by the ferromagnet/insulator interface properties [4]. In light of maintaining a nanometer-thick tunneling barrier continuous, it appears indispensable to have an ultra-smooth surface for the bottom electrode before and after annealing.

As a way to achieve ultra-smooth interfaces, we have considered employing amorphous films instead of the traditionally used Ta layers with an emphasis given on understanding underlayer effects. Amorphous materials in absence of grain boundaries, in principle, could offer better surface smoothness as well as interdiffusion resistance. Among many candidates, we have chosen CoNbZr because it has demonstrated good thermal stability [5] and smooth surface structure [6]. The main purpose of this study is to investigate the MR ratio, thermal behaviors, *I-V* and V_h behaviors of the new MTJ structures comprising CoNbZr layers.

2. Experimental Procedure

Tunnel junctions consisting of $Si/SiO_2/Co_{85.5}Nb_8Zr_{6.5}$ or Ta $2/Co_{90}Fe_{10}$ $8/Ir_{20}Mn_{80}$ $7.5/Co_{90}Fe_{10}$ 3/Al 1.6 + oxidation/ $Co_{90}Fe_{10}$ $3/Co_{85.5}Nb_8Zr_{6.5}$ or Ta 2 (in nm) were prepared by a four-target rf magnetron sputtering system under typical base pressure below 5×10^{-7} Torr. A Co target with small Nb and Zr chips added were used to get proper composition of the CoNbZr films. The film composition was occasionally confirmed by energy dispersive x-ray spectroscopy. The junctions were patterned by a set of metal shadow mask with an opening area of 200×200 μm^2. Tunnel barriers were formed by oxidizing 1.6 nm thick Al layers under rf plasma environment; flowing pure oxygen at 40 sccm to maintain 100 mTorr, and at the power density of 3.44 W/cm^2.

Annealing was done *in situ* at 300°C in 5×10^{-6} Torr vacuum under the applied field of 500 Oe. The ramp up and cool down rates were 2.5°C/s and 1°C/s, respectively.

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The temperature dependence was measured using a cryogenic dewar in the temperature range from 10 K to 300 K. The interface roughness and crystalline texture of the film were characterized by atomic force microscopy (AFM) and x-ray diffraction (XRD), respectively. To uncover interdiffusion behaviors, Auger electron spectroscopy (AES) was used.

3. Results and Discussion

The MR ratio and resistance variations of MTJ samples as a function of measuring temperature were shown in Fig. 1 for as-deposited and 300°C annealing conditions. At as-deposited state, the CoNbZr-based MTJ showed a lower MR ratio (11%) than the Ta-based MTJ (15%) measured at RT. We thought the cause of exhibiting lower ratio was due to a poorly developed crystal texture. To confirm the crystallinity of two MTJ structures, we have compared XRD patterns as depicted in Fig. 2. As clearly seen, the development of crystallinity was limited for the CoNbZr-based MTJ due to the presence of an amorphous underlayer.

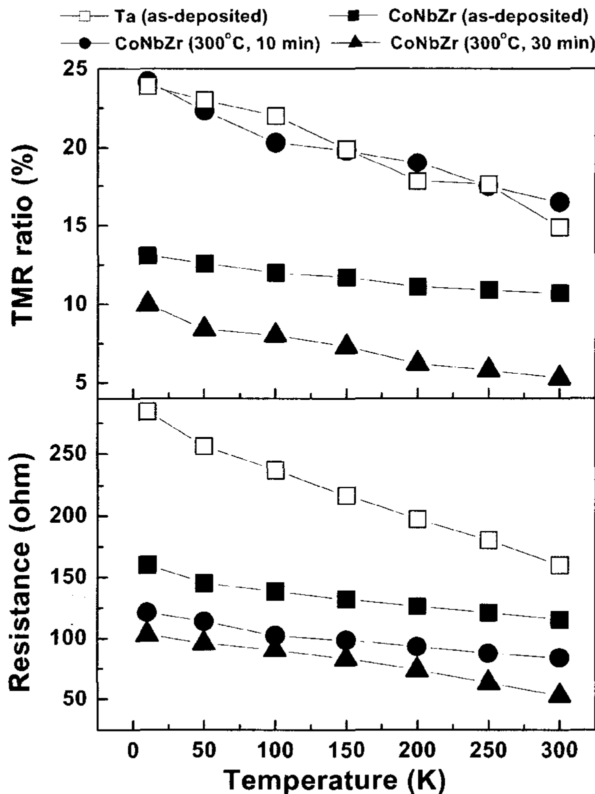

Fig. 1. TMR ratio and electrical resistance as a function of measuring temperature. The tunnel junctions consisted of Si/ SiO_2/CoNbZr or Ta 2/CoFe 8/IrMn 7.5/CoFe 3/Al 1.6 + oxidation/CoFe 3/CoNbZr or Ta 2 (in nm).

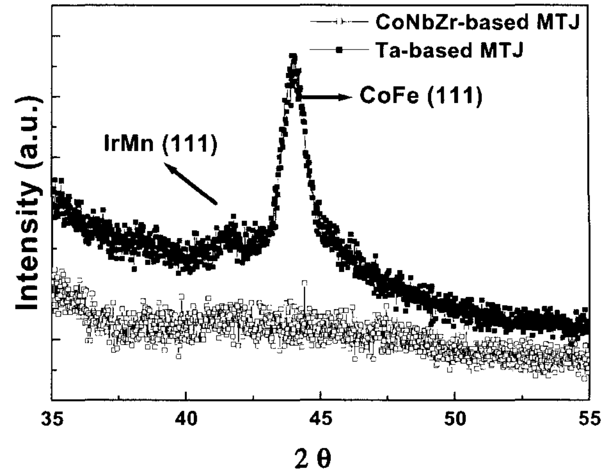

Fig. 2. XRD patterns of CoNbZr and Ta-based MTJs at as-deposited state.

When annealed (at 300°C for 10 min), MR ratio of the CoNbZr-based MTJ was increased from 11% to 17% while resistance was decreased from 115 W to 84 W. Unlike the CoNbZr-based MTJ, the Ta-based structure exhibited short-circuiting probably due to the barrier discontinuity after the same annealing condition, resulting in unmeaningful results. The increase in MR can be attributed to an improved interface with the bottom portion of ferromagnetic electrode [7]. By the long time annealing, Mn from the antiferromagnetic IrMn layer interdiffused [8] toward the CoFe pinned layer, thus the same CoNbZr sample that underwent 300°C, 30 min annealing displayed a reduction in the TMR ratio compared to the 10 min annealing one. Presumably a small amount of interdiffusion has deteriorated the MR of the 30 min annealed sample (the detailed AES data are not shown here). By using Simmons equation we could estimate the barrier height and width. By plugging these values into the following equation, we could calculate the spin polarization [9].

$$G_T(T)=G_0\frac{CT}{\sin(CT)} \tag{1}$$

$$C=1.387\frac{d}{\sqrt{\Phi}}10^6\sqrt{eV}/(mK) \tag{2}$$

The spin polarization of the both CoNbZr-based and Ta-based MTJs was shown in Fig. 3 as a function of measurement temperature. We speculated that the difference in thermal stability might be attributed to the difference in interfacial roughness, if any. To confirm our speculation, we have measured the layer surface roughness at all layer construction stages up to the alumina barriers by AFM. As displayed in Table 1, the CoNbZr-

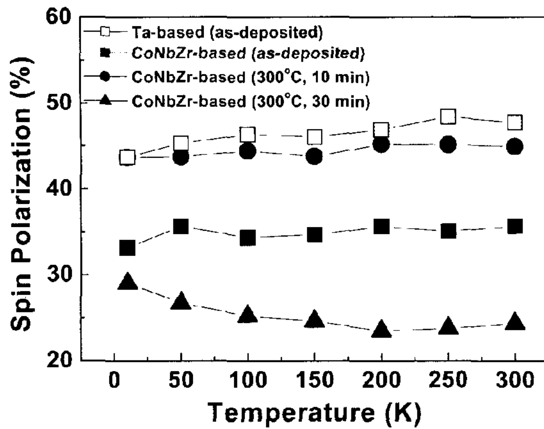

Fig. 3. Spin polarization as a function of temperature.

based bottom structure, in comparison with the Ta-based one, offered smaller root-mean-square (rms) surface roughnesses (0.12 vs. 0.26 nm). Another encouraging feature is that the surface smoothness was nearly preserved before and after annealing for the CoNbZr-based structure (from 0.12 to 0.16 nm) while that of Ta-based one became worse (from 0.26 to 0.34 nm) after annealing (at 300°C, 10 min). Therefore, achieving very smooth interfaces, in particular, up to the tunnel barrier, is beneficial for thermal stability of the MTJs.

Bias voltage dependence of the MR ratio in MTJs can be affected by several factors: metal particles, magnons, magnetic impurities, and etc [10]. In this study, we regarded interfacial roughness as a source of imperfection, and attempted to uncover the temperature-dependent bias voltage V_h (defined as a voltage where a MR ratio becomes half of its unbiased value) variation by surface roughness of the tunnel barrier. The CoNbZr-based MTJs exhibit better V_h characteristics than the Ta-based one as depicted in Fig. 4. Moreover, V_h gradually increased at higher temperatures in the CoNbZr-based MTJs whereas almost no change was observed for the Ta-based one. To confirm the surface effect on bias voltage characteristics, we have prepared a sample where the CoNbZr underlayer alone was deposited intentionally at 10 mTorr. This particular sample showed a rms roughness of 0.18 nm when measured on the surface of the tunnel barrier. At as-deposited state, the surface-roughened CoNbZr-based MTJ showed 151 mV of V_h at 10 K, which gradually

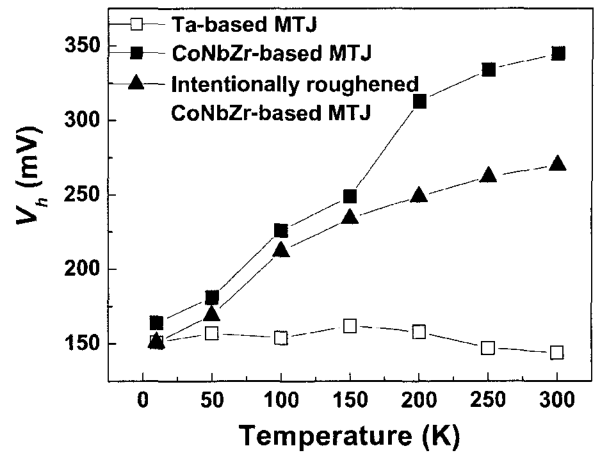

Fig. 4. V_h variation of various MTJ samples as a function of measured temperature.

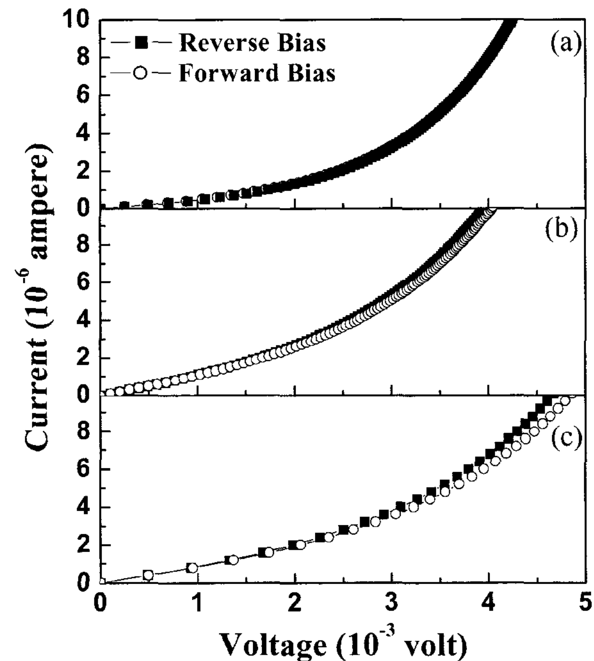

Fig. 5. The I-V curves of samples with different surface roughness values: (a) CoNbZr-based MTJ, (b) Intentionally roughened CoNbZr-based MTJ, and (c) Ta-based MTJ.

increased up to 270 mV at 300 K. The MTJs with rougher surfaces exhibited lower and less rapid increase of V_h. Thus the smoother junction has higher V_h. The I-V curves are displayed in Fig. 5. They are either symmetric or

Table 1. The evolution of rms surface roughness after subsequent layer depositions

Surface roughness (rms) As-dep./Annealed (300°C, 10 min)	Seed layer only (nm)	Up to CoFe (nm)	Up to AlOx (nm)
Ta-based MTJ	0.12 / 0.15	0.18 / 0.19	0.26 / 0.34
CoNbZr-based MTJ	0.09 / 0.11	0.10 / 0.12	0.12 / 0.16

asymmetric depending on the bias direction, say, forward (from top to bottom electrode) or reverse (from bottom to top electrode). As shown in Fig. 5(a) and 5(b), the CoNbZr-based MTJs exhibited nearly symmetric *I-V* curves. However the Ta-based MTJ showed an asymmetric curve. In rougher junctions, there might be more unoxidized residual Al and/or magnetically dead zone in the valley and summit portion of the bottom electrode, respectively [11].

Therefore, the tunnelling electrons in the rougher junction would experience more scattering events at the ferromagnet/barrier interfaces. The bias asymmetry can be attributed to an asymmetric tunnelling behavior resulting from the presence of interfacial roughness. The *I-V* curves became more asymmetric at higher voltage range for rougher junctions. Considering the forward biasing cases, the breakdown voltages of the samples (a), (b), and (c) were 1.04 V, 0.92 V, and 0.87 V at as-deposited, and 1.32 V, 1.05 V, and 0.93 V after annealing (300°C, 10 min), respectively.

4. Conclusions

Magnetic tunnel junctions (MTJs) comprising amorphous CoNbZr layers have been investigated. Unlike Ta-based tunnel junctions, CoNbZr junctions did not possess crystalline structures. At elevated temperature (300°C), a short-time (10 min) annealing promoted an increase and a decrease in MR ratio and electrical resistance, respectively, for CoNbZr-based tunnel junctions. However, a longer annealing (30 min) deteriorated MR properties likely due to interlayer diffusion. The use of a thin CoNbZr film as an underlayer resulted in ultra-smooth interfaces for the bottom electrode, which was beneficial for increasing thermal stability and increasing V_h characteristics than Ta-based MTJs. To achieve ultra-smooth interface, the use of an amorphous CoNbZr layer as an underlayer was proven effective.

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