Magnetic Tunnel Junction Based on MgO Barrier Prepared by Natural Oxidation and Direct Sputtering Deposition

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Abstract: Magnetic tunnel junctions (MTJs) based on MgO barrier have been fabricated by sputtering single crystal MgO target and metal Mg target, respectively, using magnetic sputtering system Nordiko 2000. MgO barriers have been formed by a multi-step deposition and natural oxidization of Mg layer. Mg layer thickness, oxygen flow rate and oxidization time were adjusted and the tunnel magnetoresistance (TMR) ratio of optimal MTJs is over 60% at annealing temperature 385 ℃. The (001) MgO crystal structure was obtained when the separation distance between MgO target and substrate is less than 6 cm. The TMR ratio of most MgO based MTJs are over 100% at the separation distance of 5 cm and annealing temperature 340 ℃. The TMR ratios of MTJs are almost zero when the separation distance ranges from 6 to 10 cm, due to the amorphous nature of the MgO film.

Keywords: Magnetic tunnel junctions; MgO; Crystal structure; Magnetic sputtering system

Introduction

Since Julliere's research group investigated the first spin-dependent tunneling junctions based on Co and Fe as electrode materials and GeO as insulating barrier in 1974 [1], magnetic tunneling junctions (MTJs) have been extensively studied due to its promising applications in the hard disk read heads, magnetoresistive random access memory and sensors [2,3]. The MTJs based on different tunnel barriers such as NiO [4] and Gd_2O_3 [5], showed very low tunnel magnetoresistance (TMR) effects. The TMR breakthrough is that Miyarzaki et. al. group reported MTJs based on amorphous Al_2O_3 barriers attained TMR of 18% at room temperature in 1995 [6]. At the present, the optimal MTJs based on aluminum oxide barrier can reach TMR ratio of 80% [3]. However, it is difficult for MTJs based on Al_2O_3 barrier to further improve TMR ratio. According to the first-principle electronic structure calculations, the ordered (001) oriented Fe/MgO/Fe MTJs structure were suggested to reach TMR ratio more than 100% to even 1000% [7]. This giant TMR ratio was thought to be caused by an interfacial spin-dependent electronic state with Δ_1 symmetry at the Fermi energy. Along this theoretical calculation, S. S. P. Parkin group [8] and Shinji Yuasa group [9] reported giant TMR ratio up to 220% and 180% at room temperature in 2004, respectively. The higher TMR ratio of 604% was further reported in CoFeB/MgO(001)/CoFeB MTJs using magnetic sputtering deposition [2]. To realize high TMR in MTJs, it is important to deposit (001) MgO crystallographic orientation. Wang et al reported that an improved crystallinity of the MgO (001) layer is a main reason for increasing TMR ratio at high annealing temperature [10].

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The excellent crystalline (001) MgO layer and (200) CoFeB layer are both primary factors for the higher TMR in CoFeB/MgO/CoFeB structure, confirmed by X-ray diffraction [11].

The (001) orientation MgO layer has been fabricated with different methods, such as MBE [9], magnetic sputtering deposition [8,10] and ion beam deposition [12], etc. The deposition parameters, such as Ar pressure [13], sputtering power, deposition rate and the distance between target and substrate would usually affect the crystal structure of MgO layer in magnetic sputtering systems. Therefore, the careful research work that the influence of deposition parameters on the crystal structure of MgO layer and the TMR ratio of MgO based MTJs is important for quickly and successfully fabricating (001) MgO crystal structure and obtaining higher TMR ratio of MgO based MTJs. In this study, we investigated MTJs based on MgO barrier fabricated by magnetic sputtering single crystal MgO target and metal Mg target, respectively. As for MgO barrier deposited with single crystal MgO target, the distance between MgO target and substrate is sensitive to the crystal structure of MgO layer. For MgO barrier prepared by sputtering metal Mg target, the metal Mg layer with several nanometers was naturally oxidized to form MgO barrier. The sputtering parameters of oxygen flow rate and pressure, oxidization time and Mg layer thickness apparently influence TMR ratio and the products of resistance and area (RA).

Experimental Procedures

The thin films for the MTJs were deposited onto glass substrate using a magnetron sputtering system (Nordiko 2000), with a base pressure of 5×10^{-8} Torr. A magnetic field of 20 Oe was applied to induce parallel easy axis in the bottom and top magnetic electrodes during deposition. The fundamental structure of the MTJ is Ta70/Ru50/Ta50/MnPt200/CoFe25/Ru8/CoFeB30/MgO(t_{MgO})/CoFeB30/Ta70/TiWN150 (in Å). The t_{MgO} represents the nominal thickness of MgO barriers. For sputtering metal Mg target, MgO barriers were formed by a several-step natural oxidization. First, the Mg layer was deposited on top of bottom magnetic electrode at deposition rate of 0.26 Å/s in the main chamber, and then oxidized by pure oxygen to form MgO layer in the load-lock chamber. To fully oxidize Mg layer and get right thickness of MgO barrier, the process of depositing Mg layer and natural oxidization need to be repeated several times in sequence. For sputtering single crystal MgO target, MgO barriers were deposited by RF sputtering and their nominal thickness was varied from 15 Å to 30 Å. The micro-size junctions were patterned by a self-aligned micro-

fabrication process using direct-write laser-lithography, ion-beam milling and lifting off, defining junction areas down to 1×1 um^2. Patterned samples were annealed with different temperature in vacuum pressure of 10^{-6} Torr for 1 h, and then moved into a magnetic field of 1 T while cooling down. The TMR transfer curves were measured with a four-contact DC method. The crystalline structure of MgO films was characterized by X-ray diffractometer (XRD, Siemens D-5000, molybdenum source Kα=0.7107 Å) [12].
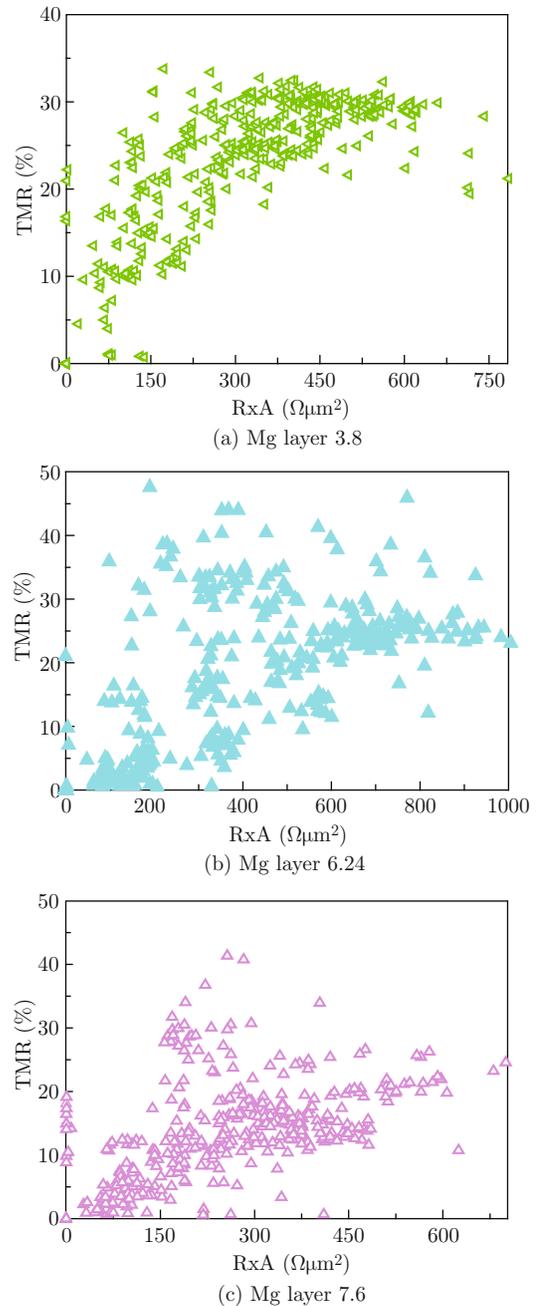

Fig. 1   TMR ratios versus RA products for MTJs with different Mg layer thickness.

Results and Discussion

Figure 1 shows TMR ratios versus RA products with different Mg layer thickness. The Mg layer thickness per step deposition was chosen 3.8 Å, 6.24 Å and 7.6 Å, respectively. MTJs were annealed at 370 ℃ for 1 hour. Data are from ~324 MTJs assemblage. The total nominal thickness of Mg layer was about 30 Å. The first Mg layer was oxidized for 40 seconds and the following several Mg layers were oxidized for 80 seconds at 2 sccm oxygen flow rate. The shortened oxidization time of the first step is to avoid oxidizing the bottom electrode, which can decrease TMR of MTJs [14]. It seems difficult to judge which depositing Mg layer thickness is better for higher TMR ratio, as illustrated in Fig. 1. However, MTJs with 6.24 Å Mg layer are probably worthy further optimization because that the number of MTJs with Mg layer 6.24 Å of more than 40% TMR is more than that of MTJs with Mg layer 7.6 Å and 3.8 Å.

For MTJs with Mg layer 6.24 Å, TMR ratio versus RA products for different oxidization time was shown in Fig. 2. The oxygen flow rate of oxidizing Mg layer was 16 sccm. The MTJs were annealed at 370 ℃ for 1 h. The average RA products of MTJs with oxidization time 80 and 100 seconds are apparently larger than

that of MTJs with 60 seconds due to the complete oxidization of these Mg layers [15]. However, the higher TMR ratios of MTJs with oxidization time 60, 80 and 100 seconds are all over 40% at annealing temperature 370 ℃. The TMR ratio of MTJs was further improved and obtained more than 60% at annealing temperature 385 ℃, as shown in Fig. 2(d).

For MgO barriers deposited by single crystal MgO target, it is very important for high TMR ratio to obtain (001) MgO crystal structure [7,9]. Here, sputtering parameters, such as RF power, Ar gas flow, gas pressure and the separation distance between MgO target and glass substrate were optimized. However, our experiments indicated that the separation distance between MgO target and substrate is most important parameter in magnetic sputtering system Nordiko 2000. Fig. 3 shows the XRD scan curves of MgO films under different sputtering condition. When the separation distance is between 5 cm and 6 cm, the XRD peak of MgO (002) orientation appears at ~19.4° [12]. However, the XRD peak of MgO film with separation distance 6 cm is weaker compared to those of separation distance 5 cm. MgO films are almost amorphous at the separation distance ranging from 6 to 10 cm. These results showed that the crystal status of MgO layer is sensitive to the separation distance. When the separation
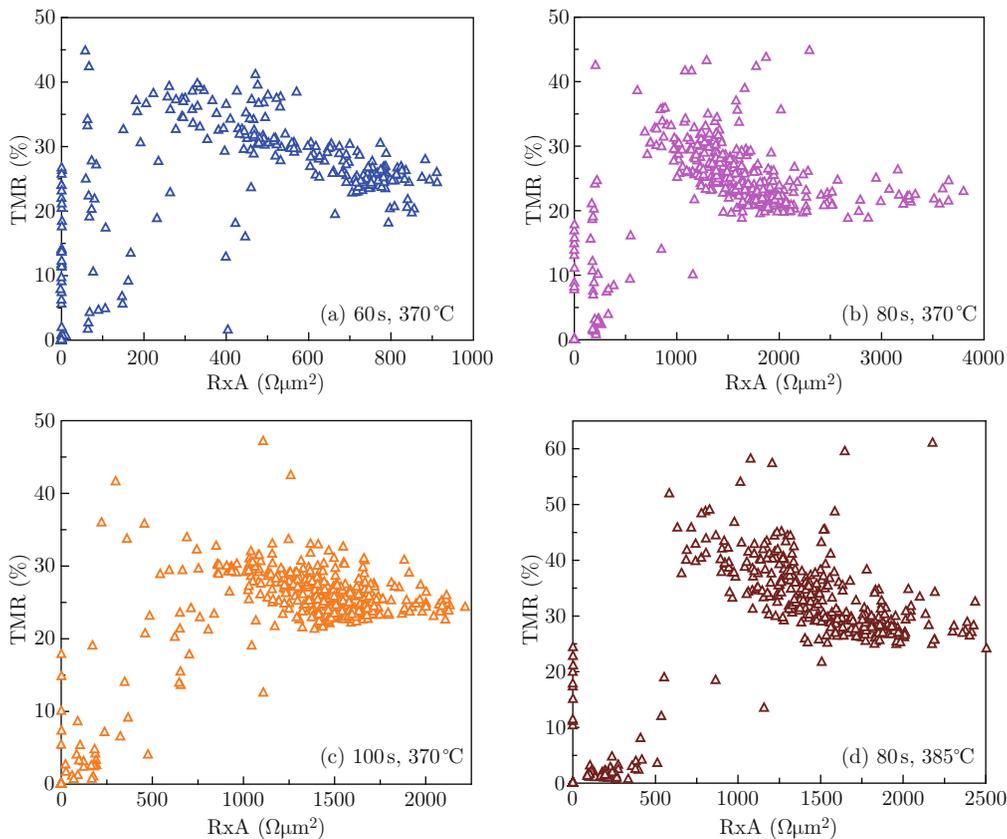

Fig. 2   TMR ratios versus RA products for MTJs with different oxidization time and annealing temperature.
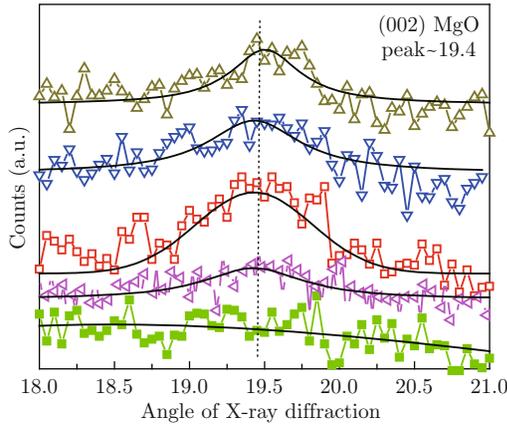
Fig. 3   X-ray diffraction scan curves of MgO films. Solid lines are fitting curves. The deposition conditions of MgO film: (□) RF power 170 W, Ar pressure 12 mTorr, separation distance 5 cm; (■) 170 W, 12 mTorr and 9.5 cm; (▽) 170 W, 5 mTorr and 5 cm; (△) 170 W, 8 mTorr and 5 cm; (◁) 170 W, 12 mTorr and 6 cm.

distance was set 5 cm, the (002) MgO crystal structure was easily formed in the large range of RF power and Ar gas pressure.

Figure 4 shows the TMR curve of the representative MTJs. MgO barriers were deposited at the separation distance 5 cm. The MTJs were annealed at 340 °C for 1 h. The MTJ MgO barrier thickness, RF sputtering power and Ar pressure were 19 Å, 150 W and 12 mTorr (Fig. 4(a)), 15 Å, 150 W and 12 mTorr (Fig. 4(b)), and 15 Å, 100 W and 8 mTorr (Fig. 4(c)), respectively. The TMR ratios of MTJs are about 130% (Fig. 4(a)), 150% (Fig. 4(b)) and 170% (Fig. 4(c)), respectively. One sample with one kind of MgO barrier deposition condition can be fabricated ~324 MTJs. Here, we just chose a representative MTJ to plot a TMR curve under one deposition condition. From a statistical point of view, TMR ratios of most MTJs with above three kinds of MgO barrier deposition conditions are more than 100%. Our results demonstrated that TMR ratios of most MTJs are always more than 100% for MgO barrier deposited with RF sputtering power ranging from 100 W to 200 W, and Ar pressure ranging from 5 mTorr to 20 mTorr at the separation distance of 5 cm. However, TMR ratios of MTJ are almost zero when the separation distance is larger than 6 cm due to amorphous MgO films. Therefore, the separation distance between MgO target and glass substrate is most important parameter to get high TMR ratio of MgO based MTJs in magnetic sputtering system Nordiko 2000.

Conclusions

In summary, MgO barriers were investigated by sputtering single crystal MgO target and natural oxidization, respectively. Mg layer thickness, oxidation time and oxygen flow rate are relative to TMR ratios under
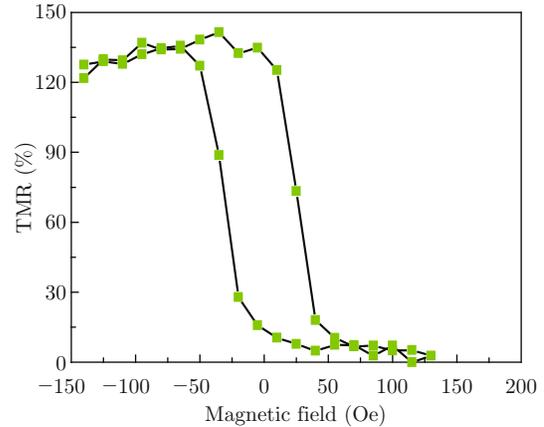

(a) RF 150 W, 8 mTorr, MgO 19 Å
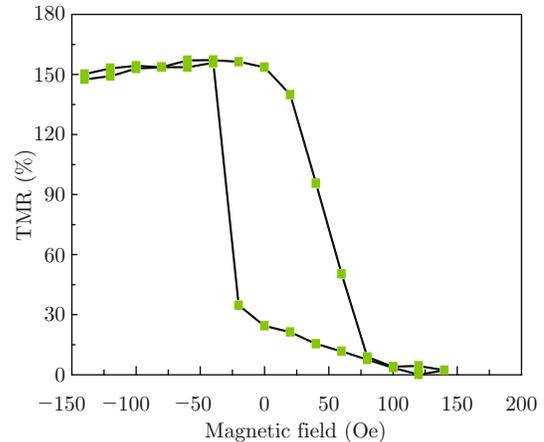
(b) RF 150 W, 12 mTorr, MgO 15 Å
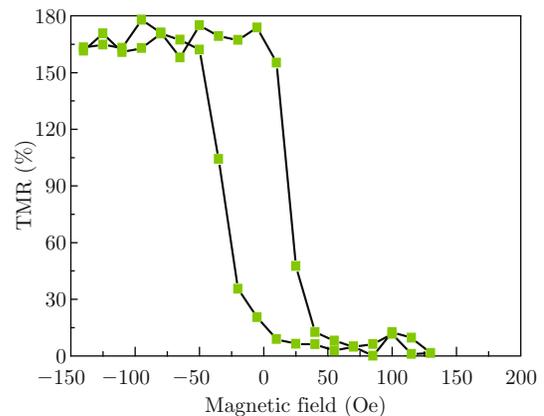
(c) RF 100 W, 12 mTorr, MgO 15 Å

Fig. 4   TMR curves of MgO barriers deposited with different deposition parameters.

natural oxidization method. The TMR ratio of the optimal MTJ is more than 60% at annealing temperature 385 °C. The separation distance of MgO target and substrate is more important for depositing (001) MgO crystal structure in magnetic sputtering system Nordiko 2000. The TMR ratios of reasonable MTJs are more than 100% at the separation distance of 5 cm. The MgO film is almost amorphous when the separation distance varies from 6 to 10 cm.

Acknowledgments

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