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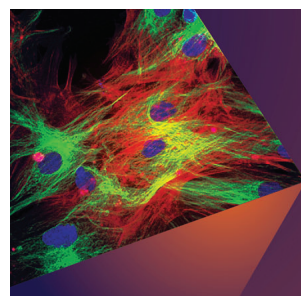
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Ultralow resistance-area product of $0.4 \Omega(\mu\text{m})^2$ and high magnetoresistance above 50% in CoFeB/MgO/CoFeB magnetic tunnel junctions

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An ultralow resistance-area (RA) product of $0.4 \Omega(\mu\text{m})^2$ was achieved in CoFeB/MgO/CoFeB magnetic tunnel junctions with a high magnetoresistance ratio of 57% at room temperature. Various growth conditions for polycrystalline MgO(001) tunneling barrier were optimized to improve the crystalline orientation of the MgO(001) layer, which resulted in a significant enhancement of magnetoresistance in an ultralow RA region below $1 \Omega(\mu\text{m})^2$. Removal of residual H_2O molecules from a growth chamber was especially effective in improving the crystalline orientation. The present achievements will enable the development of highly sensitive read heads for ultrahigh-density hard disk drives. © 2006 American Institute of Physics. [DOI: 10.1063/1.2352046]

A highly sensitive magnetic sensor is indispensable for ultrahigh-density hard disk drives. Current-perpendicular-to-plane magnetic sensors (read heads) have been proposed for areal recording densities beyond 100 Gb/in.².¹⁻⁴ Among them, the tunneling magnetoresistive (TMR) head with a magnetic tunnel junction (MTJ) is the most promising candidate. For the next-generation read head, a high magnetoresistance (MR) ratio is required to compensate for the reduction in the signal-to-noise ratio due to the shrinking recording bit size, and a low resistance-area (RA) product is required to satisfy the impedance matching indispensable for high-speed readout.^{3,5} TMR heads with amorphous Al-O tunneling barriers developed for recording densities above 100 Gb/in.² have RA products of $2-5 \Omega(\mu\text{m})^2$ with MR ratios of about 20% at room temperature (RT).^{6,7} However, the demand for higher recording densities requires a MR ratio significantly higher than 20% as well as an even lower RA product.

MTJs with a crystalline MgO(001) tunneling barrier have been developed for magnetoresistive random access memory. The MgO-based MTJs exhibit MR ratios much higher than the Al-O-based MTJs.⁸⁻¹¹ This giant TMR effect originates from coherent spin-dependent tunneling of highly-spin-polarized Δ_1 states.^{12,13} In our previous study,¹⁴ we achieved a low RA product of $2.4 \Omega(\mu\text{m})^2$ with a giant MR ratio of 138% at RT in CoFeB/MgO/CoFeB MTJs with a (001)-oriented polycrystalline (textured) MgO tunneling barrier. Although these properties are sufficient for a recording density of about 200 Gbit/in.², an ultralow RA product below $1 \Omega(\mu\text{m})^2$ with a MR ratio above 50% is required for recording densities beyond 200 Gbit/in.² In the previous study, however, the MR ratio was observed to decrease

sharply as the RA product approached $1 \Omega(\mu\text{m})^2$, and the TMR effect almost disappeared when $RA < 1 \Omega(\mu\text{m})^2$.¹⁴

The TMR effect in MgO-based MTJs is sensitive to the crystalline orientation of the MgO barrier. The (001) orientation of MgO barrier is indispensable for the giant TMR effect because the four-fold symmetry in the barrier and at the barrier/electrode interfaces is essential for the coherent tunneling of Δ_1 electrons.^{12,13} In the CoFeB/MgO/CoFeB MTJs, the MgO layer deposited on the amorphous CoFeB bottom electrode layer has an amorphous structure in the initial growth stage and begins to crystallize in the (001) orientation when the MgO thickness (t_{MgO}) exceeds about 1.0 nm.¹⁵ This is considered to be the principal mechanism of the sharp drop in the MR ratio in an ultrathin MgO barrier region (i.e., an ultralow RA region). Recently, Yuasa *et al.* reported an ultralow RA of $0.4 \Omega(\mu\text{m})^2$ with a relatively high MR ratio of 30% at RT in fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs.¹⁶ Even though epitaxial MTJs are not useful for applications, this result is notable because it proved that a highly oriented epitaxial MgO(001) barrier was effective in enhancing the MR ratio in the ultralow RA region. Another recent development, from our previous study,¹⁴ is the discovery that insertion of a 0.4 nm thick Mg metal layer between the CoFeB bottom electrode and the MgO barrier is effective in improving the crystalline orientation of MgO(001) and consequently in enhancing the MR ratio in a low RA region of a few $\Omega(\mu\text{m})^2$. In this method, however, a partial oxidation of the Mg metal layer makes it difficult to reduce the effective thickness of MgO(001) to about 1.0 nm, which is required to achieve a RA below $1 \Omega(\mu\text{m})^2$. In this study, we developed a new process for growing an ultrathin highly oriented MgO(001) layer without inserting the Mg metal layer and succeeded in reducing RA to $0.4 \Omega(\mu\text{m})^2$ while keeping a MR ratio above 50% at RT.

We deposited the thin films for the MTJs onto a thermally oxidized Si(001) wafer using a magnetron sputtering system (Canon ANELVA C-7100) with a base pressure on

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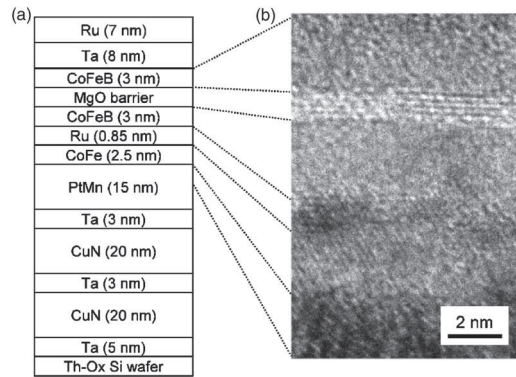


FIG. 1. (a) Cross-section structure of CoFeB/MgO/CoFeB magnetic tunnel junction (MTJ). (b) High-resolution cross-section transmission electron microscopy (TEM) image of CoFeB/MgO/CoFeB MTJ with designed MgO thickness of 1.02 nm.

the order of 10^{-9} Torr. As shown in Fig. 1(a), the fundamental structure of the MTJs was a substrate/seed layer/PtMn(15) / Co₇₀Fe₃₀(2.5) / Ru(0.85) / CoFeB(3.0) / MgO / CoFeB(3.0)/cap layer (thickness in nanometers). The seed and cap layers were Ta(5)/CuN(20)/Ta(3)/CuN(20)/Ta(3) and Ta(8)/Cu(30)/Ta(5)/Ru(7) (thickness in nanometers), respectively. In this study, we deposited the MgO barrier layer directly on the CoFeB bottom electrode layer without an insertion of Mg metal layer. Designed thickness of the MgO barrier, defined as a product of deposition rate and deposition time, varied from 1.01 to 1.12 nm. The actual thickness of the MgO layer as observed by high-resolution cross-section transmission electron microscopy (TEM) was in good agreement with the designed thickness, as shown later. All the metallic layers were deposited using a dc magnetron sputtering method. The insulating MgO layer was deposited by rf sputtering directly from a sintered MgO target having a purity of 99.99% and a density of 95 vol %. The CoFeB layers were deposited using a Co₆₀Fe₂₀B₂₀ (at %) alloy target. The Ar gas pressure during sputtering was kept low (about 0.2 mTorr), except for the deposition of the MgO layer, which was carried out at various Ar pressures. To achieve a better crystalline orientation of the MgO(001) layer, we carefully optimized various conditions for the growth of MgO such as the Ar gas pressure and base pressure of the sputtering chamber. The MTJ films were annealed in a high-vacuum furnace at 360 °C for 2 h in a magnetic field of 8 kOe to enhance the exchange bias field of the pinned layer as well as to crystallize the CoFeB electrodes. Then, the films were

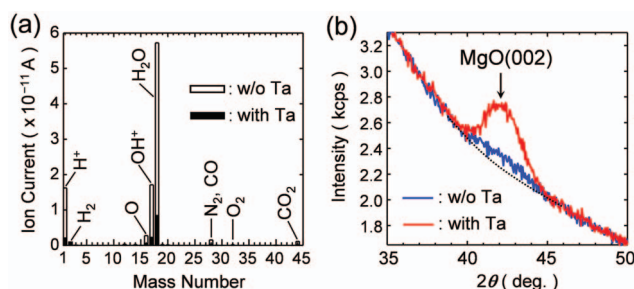


FIG. 2. (Color) (a) Quadrupole mass spectra of sputtering chamber with (black bars) and without a Ta getter (white bars). (b) X-ray diffraction profiles from 3 nm thick MgO layers prepared with (red line) and without Ta getter (blue line). The dotted line represents background intensity drawn as a guide for the eyes.

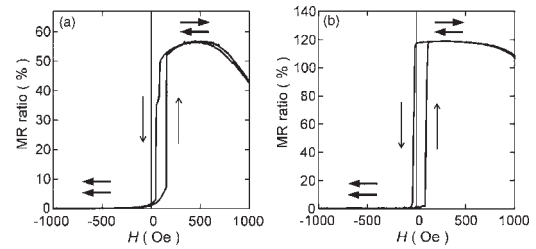


FIG. 3. Magnetoresistance curves at room temperature for optimized CoFeB/MgO/CoFeB MTJs. (a) Designed MgO thickness=1.02 nm, RA=0.4 Ω(μm)², and MR ratio=57%. (b) Designed MgO thickness=1.05 nm, RA=1.1 Ω(μm)², and MR ratio=119%. The thick arrows represent magnetization alignments.

fabricated into deep-submicron-sized MTJs by using electron-beam lithography and Ar-ion milling. The actual lateral sizes of MTJs were slightly larger than the designed size (the size of the electron-beam resist patterns) because the sidewalls of the MTJs were tapered after the Ar-ion milling. We estimated the actual MTJ sizes by using scanning electron microscopy observations after the Ar milling. For the designed MTJ size of 50×150 nm², for example, the actual junction size was estimated to be 70×170 nm². We confirmed a linear relationship between the junction resistance (R) and an inverse of the estimated junction area ($1/A$) using MTJs with various junction areas, which proves the validity of the estimated A . The MR ratio and RA product were measured using the dc four-point probe method. We also measured x-ray diffraction profiles (θ - 2θ scan) of MTJ films to evaluate the degree of MgO(001) crystalline orientation. Because the MgO barriers of the ultralow resistance MTJs were too thin (~ 1 nm) to be detected by x-ray diffraction (XRD) measurements, we also prepared MTJ films with a much thicker MgO layer ($t_{\text{MgO}}=3\text{--}10$ nm) just to measure the XRD profiles. The intensity of the MgO(002) diffraction peak was used to evaluate the degree of (001) orientation.

The MR ratio was observed to be sensitive to Ar gas pressure (P_{Ar}) during the MgO deposition. The Ar gas was introduced from the top part of the sputtering chamber, where the sputtering cathodes were located. The Ar gas pressure was measured with a capacitance manometer gauge attached on the bottom part of the chamber, where the substrate and a cryopump were located. Therefore the measured pressure is the Ar gas pressure around the substrate. The Ar gas pressure was controlled by a flow rate of the Ar gas. The sputtering deposition of MgO was carried out at $P_{\text{Ar}}=0.23$, 0.52, and 0.75 mTorr. The (002) peak intensity of XRD from a 10 nm thick MgO layer was observed to increase with increasing P_{Ar} . Consequently, the MR ratio increased with P_{Ar} . Because 0.75 mTorr was the highest possible Ar gas pressure in the sputtering system, we adopted an Ar gas pressure of 0.75 mTorr for optimized growths. The growth of the MgO layer was also found to be sensitive to the base pressure of the sputtering chamber. The partial pressure of residual H₂O molecules was found to be an especially important factor. To remove the residual H₂O molecules, we used a tantalum (Ta) getter in the following way. Just before the deposition of MgO, Ta was sputtered from another sputtering cathode installed in the same growth chamber. This Ta layer was not deposited on the sample substrate but on other parts of the chamber such as the inner sidewalls. The Ta layer acted as a getter to adsorb residual H₂O molecules. Typical

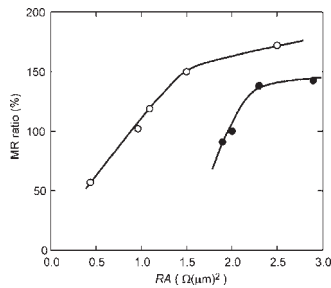


FIG. 4. MR ratio at room temperature for CoFeB/MgO/CoFeB MTJs as a function of RA product. The open circles represent optimized results of this study. The solid circles represent results of our previous study (Ref. 14).

quadrupole mass spectra of the sputtering chamber with and without the Ta getter are shown in Fig. 2(a). Without the Ta getter, the main residual gas was H_2O . The Ta getter was effective in reducing the partial pressure of H_2O . Typical XRD profiles from 3 nm thick MgO layers with and without the Ta getter are shown in Fig. 2(b). The (002) diffraction peak of the 3 nm thick MgO layer without the Ta getter was only faintly detected by XRD because of its weak intensity. When the MgO layer was deposited with the Ta getter, on the other hand, the (002) peak was clearly observed because of the enhanced peak intensity. The use of the Ta getter resulted in about a threefold increase in the MR ratio in the ultralow RA region. It is speculated that H_2O molecules adsorbed on the surface of amorphous CoFeB prevent the textured growth of MgO(001) in the initial growth stage. It should be noted here that the removal of H_2O molecules by the Ta getter might also have resulted in the suppression of overoxidation at the barrier/electrode interfaces, which is also known to be effective in enhancing the TMR effect.^{9,17}

After these experiments, we adopted an Ar gas pressure of 0.75 mTorr with the Ta getter as the optimum conditions. A typical cross-section TEM image of the optimized MTJ is shown in Fig. 1(b). The crystalline lattice image of the textured MgO(001) barrier is clearly identified in the TEM image. About five monoatomic layers of MgO(001) are identified in the TEM image. This corresponds to a MgO thickness of about 1.0 nm. Note that this value is in good agreement with the designed MgO thickness, which was 1.02 nm. Typical magnetoresistance curves of the optimized MTJs are shown in Figs. 3(a) and 3(b). An ultralow RA of $0.4 \Omega(\mu m)^2$ was achieved in the MTJ having a high MR ratio of 57% at RT. Figure 4 shows the plots of the MR ratio at RT as a function of RA product, together with the results of our previous study.¹⁴ The optimizations of the MgO growth conditions resulted in the significant enhancement of MR ratios especially in the ultralow RA region below $1 \Omega(\mu m)^2$. In addition to achieving a $RA=0.4 \Omega(\mu m)^2$ with a MR ratio = 57%, we also achieved a $RA=0.96 \Omega(\mu m)^2$ with a MR ratio = 102%, and a $RA=1.1 \Omega(\mu m)^2$ with a MR ratio = 119%. It should be noted that these MR ratios are much higher than those in the current-perpendicular-to-plane-giant magnetoresistance (CPP-GMR) devices using the nano-oxide layer process, in which the MR ratio is slightly higher than 8% at RA of $0.5 \Omega(\mu m)^2$,¹⁸ or in the CPP-GMR devices using the Heusler alloy as the ferromagnetic layers, in which the MR ratio is slightly higher than 10% at RA of $0.1 \Omega(\mu m)^2$.¹⁹ The results of this study satisfy the require-

ments for the ultrahigh recording densities beyond 200 Gbit/in.².

In conclusion, we optimized various growth conditions for the MgO barrier in CoFeB/MgO/CoFeB MTJs to improve the crystalline orientation of MgO(001). The optimizations resulted in the significant enhancement of MR ratio in the ultralow RA region below $1 \Omega(\mu m)^2$. An ultralow RA of $0.4 \Omega(\mu m)^2$ was achieved in the MTJ while retaining a high MR ratio of 57% at RT. These achievements will enable the development of highly sensitive read heads for ultrahigh-density hard disk drives.

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