Surface smoothing process for high-performance MgO-based magnetic tunnel junctions

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Received October 17, 2018; accepted December 4, 2018; published online January 10, 2019

We developed a surface smoothing technique for stacked Ta–B and Co–Fe–B amorphous layers in a magnetic tunnel junction (MTJ) to yield a higher magnetoresistance (MR) ratio and stronger synthetic anti-ferromagnetic (SAF) coupling. The technique yielded an ultra-smooth surface (Ra ~ 0.1 nm) for both amorphous films and gave rise to atomically flat interfaces at the subsequently deposited layers. Top-pin MTJs fabricated with the technique exhibited superior properties, such as an MR ratio up to 315% and strong SAF coupling exceeding 0.7 erg cm⁻². The smoothing process for amorphous layers will be important for fabricating the MTJs of high-performance spintronics devices.

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Magnetic tunnel junctions (MTJs) with high magnetoresistance (MR) ratios are essential for high-performance magnetoresistive random access memory (MRAM), spin-torque oscillators, and magnetic field sensors. So far, (001)-oriented MgO has been utilized as a barrier material in MTJs for its outstandingly high MR ratio, low resistance-area (RA) product, and feasible manufacturing process using sputtering.2–16) It is widely recognized that coherent tunneling of Δ₁ Bloch states is essential for a high MR ratio when the MgO/bcc Fe-based electrode has the (001) crystal orientation.17,18) In terms of the MR ratio, 300% at room temperature is a criterion for high-performance MTJs; in particular, sputter-deposited Co–Fe–B/MgO/Co–Fe–B MTJs with an exchange-biased synthetic anti-ferromagnetic (SAF) reference structure have rarely exceeded this value, except in a limited number of reports.19,20) Three technical factors are known to be important for increasing the MR ratio: the spattering conditions of the MgO deposition,21–24) the composition of Co–Fe–B,20,25) and the post-annealing temperature.16,19) In this study, we focused on a new factor for achieving a high MR ratio, the smoothness of the layer surfaces. We have developed a surface smoothing technique to modify rough stacked layers in Co–Fe–B/MgO-based MTJs into smooth ones, with the aim of attaining an MR ratio exceeding 300%. So far, there have been only a few reports on the relationship between surface roughness and the MR ratio.26,27) However, development of this technique is of particular importance, because the roughness directly determines the degree of crystal orientation; thus, greater flatness would lead to more efficient coherent tunneling and a high MR ratio.

In this study, we developed a smoothing technique for amorphous Ta–B layers stacked under Co–Fe–B/MgO/Co–Fe–B layers. We expected that such amorphous layers would be suitable for the smoothing technique because of the absence of crystal grains that might cause surface roughness. Besides the Ta–B buffer layer, we additionally attempted to smooth Co–Fe–B layers that were stacked above the MgO barrier to improve the strength of SAF coupling of a top-pin type reference (top-ref) structure.

Multilayered films including MTJ stacks were fabricated on thermally oxidized Si coupons by using a mass-production physical vapor deposition (PVD) apparatus (C-7100, Canon-Anelva). The MgO barrier layer was radio-frequency sputter-deposited using a sintered MgO target with an Ar gas pressure of about 6 × 10⁻² Pa. The RA product of MTJs were varied by the duration of the MgO sputter-deposition. The smoothing process was developed from a conventional reverse sputtering method that works in a vacuum chamber generally equipped with a C-7100. The reverse sputtering conditions were as follows: 0.005 nm s⁻¹ etching rate for both the Ta–B and Co–Fe–B films with 50 W of the input power for an 8 inch wafer and 0.1 Pa of Ar gas pressure. Surface roughness was evaluated by atomic force microscopy (AFM). The MR ratio and RA product of the MTJs were evaluated using a current-in-plane tunneling technique after post-annealing the stacks at various temperatures (Tₐ) in an external magnetic field (H) of 1 T. The magnetization (M) of the films was measured using a vibrating sample magnetometer. The microstructure of the films was observed by high-resolution transmission-electron microscopy (HR-TEM).

First, we tried to smooth the surface of a 5 nm thick Ta₅₀B₅₀ layer by using the reverse sputtering method. Figure 1(a) shows the surface roughness (Ra and RMS) as a function of the reverse sputtering duration (tₐ). The results reveal that the relatively rough initial surface with Ra = 0.18 nm and RMS = 0.22 nm [see the AFM image in Fig. 1(b)] became smooth as the duration increased, and an atomically flat surface with Ra = 0.10 nm and RMS = 0.13 nm was achieved at tₐ = 100 s [see the AFM image in Fig. 1(c)]. Furthermore, the window for the best flatness was wide, from 100 to 200 s. Such a wide window will be an advantage for the manufacturability of practical devices, because it tolerates variations in the initial roughness and areal dispersion of the etching rate. Concerning the smoothing mechanism for the Ta–B surface, we considered the following scenario during the reverse sputtering: a small input power density (50 W/8 inch) cannot take all the etched Ta–B clusters away from the surface, and some etched clusters (that may be amorphous phase) are again deposited on the surface for the downward PVD configuration. Assuming such a stirring-like process, a very small etching depth for only 0.5 nm (tₐ = 100 s) and a uniform etching rate...
on the whole surface would reasonably yield an atomically flat surface. We expected that if a reverse sputtering is done for a poly-crystalline film, an irregular etching rate for each crystal grain and boundary may not bring a smoother surface.

After the surface of the Ta–B layer was smoothed, an in-plane MTJ stack was subsequently deposited as follows: smoothed Ta$_{80}$B$_{20}$ (~5)/Ta(3)/Co$_{42}$Fe$_{36}$B$_{22}$(3)/MgO/smoothed Co$_{32}$Fe$_{36}$B$_{22}$(3)/W(0.15)/Co$_{32}$Fe$_{36}$(0.8)/Ru (0.85)/Co$_{32}$Fe$_{36}$(2.5)/Ir–Mn (10)/electrode (thickness in nm). The full stack of the MTJ is schematically shown in Fig. 2(a). Note that the Co$_{32}$Fe$_{36}$B$_{22}$ layer deposited over the MgO barrier was also smoothed [Smoothing-B process in Fig. 2(a)] to obtain a strong SAF coupling. The details of the Smoothing-B process will be mentioned later. Figure 2(b) plots the MR ratio vs. the RA product range of MTJs at $T_a = 380 \degree C$. The MR ratio for the MTJs with the smoothed Ta–B layer ($t_A = 100 s$) was much superior to that of the un-smoothed layer over the whole RA product range. We attained an MR ratio exceeding 300% (MR[RA] = 315% | 71 $\mu$Ωm$^2$). We consider that such a high MR ratio is due to the smoothed Ta–B layer improving the flatness of the Co–Fe–B/MgO/Co–Fe–B part. For the ultra-low RA product range, the smoothing process is also effective for increasing the MR ratio; i.e. an MR of 215% was achieved at an RA product of only 1.9 $\mu$Ωm$^2$. Note that for the RA product range of <1.7 $\mu$Ωm$^2$, the trend of the MR ratio was reversed with regard to the post-annealing temperature [Fig. 2(c)]: the MR ratios at $T_a = 360 \degree C$ were higher than those at 380 °C. The mechanism of reversal has not been elucidated yet. Figure 3 shows an HR-TEM image of a typical MTJ at $T_a = 380 \degree C$, which exhibited an MR[RA] ratio of 315% | 71 $\mu$Ωm$^2$. The image shows a clear out-of-plane crystalline orientation and very little roughness in the Co–Fe–B/MgO/Co–Fe–B part. A nearly perfect (001)-oriented MgO barrier is essential for obtaining a high MR ratio. Although the Ta–B/Ta interface is not visible, the image reveals that the smoothing process for the Ta–B resulted in the subsequently deposited layers, such as the Co–Fe–B and the MgO, having outstanding flatness.

Next, we examined the smoothing for the Co–Fe–B layer. Despite the stack being fabricated on the smoothed Ta–B buffer layer, roughness unavoidably arises at the Co–Fe–B layer when it overlies the MgO layer. The reason is that the surface energy of the MgO(001) surface is much lower than that of the subsequently deposited Co–Fe–B layer, resulting in the Co–Fe–B layer having poor wettability. We fabricated the sample for the smoothing test as follows: substrate/Ta (5)/MgO (15)/Co–Fe–B (5). The top surface was smoothed using the reverse sputtering method under the same conditions as the Ta–B smoothing. The flatness was evaluated by AFM. Figure 4(a) plots the roughness as a function of the reverse sputtering duration ($t_b$). We obtained the best flatness (less than 0.1 nm of Ra and RMS) for a wide range of $t_b$ from 150 to 300 s. The results showed a similar trend as those for Ta–B and revealed that the reverse sputtering was also effective at smoothing the Co–Fe–B surface. We derived a reasonable $t_b$ of 150 s and fabricated a full MTJ stack, as schematically shown in Fig. 2(a). Figures 4(b) and 4(c) are $M$–$H$ loops for the MTJs with $t_b = 0$ (un-smoothed Co–Fe–B) and 150 s (smoothed Co–Fe–B), respectively. The index $H_{ex}$ denotes the SAF-coupled exchange field. There is a considerable difference between the two loops in the characteristics of the interlayer exchange coupling (IEC) in the reference structure. For the MTJ with the smoothed Co–Fe–B, $H_{ex}$ (4.5 kOe) is much larger than that for the un-smoothed MTJ (2.7 kOe). The IEC coupling energy density ($J_{ex}$) was evaluated using...
\[ J_{ex} = H_{ex} M_{s1} t_1 M_{s2} t_2/(M_{s1} t_1 + M_{s2} t_2) \]

where \( M_{SN} \) is the saturation magnetization and \( t_N \) is the thickness for the lower \((N = 1)\) and upper \((N = 2)\) layers of the SAF structure.\(^{28}\) It was 0.79 and 0.47 erg cm\(^{-3}\) for the smoothed [Fig. 4(c)] and un-smoothed [Fig. 4(b)] cases. The values of \( J_{ex} \) reported in the previous studies are up to 0.7 erg cm\(^{-3}\) for a Ru spacer at Fig. 2. (Color online) (a) Schematic illustration of an in-plane magnetic tunnel junction (MTJ). (b) Magnetoresistance (MR) ratio as a function of resistance-area (RA) product for MTJs with un-smoothed \((t_A = 0\, s)\) and smoothed \((t_A = 100\, s)\) Ta–B layer. (c) Magnified plots of MR as a function of RA product for \( t_A = 100\, s \) post-annealed at 380 °C and 360 °C.

Fig. 3. (Color online) Cross-sectional high-resolution transmission-electron-microscopy (HR-TEM) image of the MTJ with the smoothed Ta–B layer post-annealed at 380 °C. This stack exhibited the highest magnetoresistance ratio (315%).

Fig. 4. (Color online) Results for Co–Fe–B surface smoothing. (a) Surface roughness as a function of the reverse sputtering duration \((t_B)\). (b) and (c) Magnetization loops for MTJs without smoothed \((t_B = 0\, s)\) and with best smoothed \((t_B = 150\, s)\) Co–Fe–B layer, respectively, in the top-reference structure. The index \( H_{ex} \) denotes the synthetic anti-ferromagnetic coupled exchange field.
the so-called second peak for the spacer thickness in the plot of $J_{ex}$ vs. Ru thickness (about 0.85 nm). Therefore, our MTJ with the smoothed Co–Fe–B reached almost the ideal $J_{ex}$. On the other hand, for the MTJ with the un-smoothed Co–Fe–B, the deterioration in IEC strength was probably due to the roughness at the Ru spacer. Comparing the two samples, one can conclude that the smoothing for Co–Fe–B brings out the full potential of the IEC of the SAF structure through improving the flatness at the Ru spacer. The HR-TEM image in Fig. 3 indicates the Ru layer has good flatness.

In summary, we developed a surface smoothing process for an amorphous Ta–B underlayer and Co–Fe–B electrode layers and studied its effect on the MR ratio and the magnetic properties of the SAF structure. We obtained atomically flat surfaces with $Ra \sim 0.1$ nm for both the Ta–B and Co–Fe–B layers with a wide optimum window in terms of process time. We found that smoothing the Ta–B layer yielded larger MR ratios than those of the un-smoothed case for the whole RA product range and an MTJ with an exchange-biased SAF reference structure obtained an MR ratio exceeding 300%. We also found that the smoothing for the Co–Fe–B layers achieved a strong IEC in a top-ref-type Ru-based SAF structure. The magnetic behavior of the SAF structure indicated that the smoothing brought out the full potential of the IEC ($J_{ex} > 0.7 \text{ erg cm}^{-2}$) by improving the flatness at the Ru spacer. The smoothing process reported here is very simple and beneficial for achieving a high MR ratio and strong SAF coupling. It will be a key to building spintronics devices such as magnetic sensors and MRAMs.

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