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Nitrogen effects on lowering specific junction resistance and suppressing Mn diffusion in a magnetic tunnel junction

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We investigated the annealing effects of a magnetic tunnel junction on the Fe bottom pinned layer, of which the top surface was exposed to nitrogen plasma for a few tens of seconds prior to the deposition of the Al layer. The nitrogen-treated junction shows a lower magnetoresistance (MR) ratio and specific junction resistance (RA) than the untreated normal junction. However, after annealing at 230 °C, the MR ratio of the nitrogen-treated junction recovers to optimal values of the normal junction, while the RA remains lower than that of the normal junction. Furthermore, the nitrogen-treated junction shows less reduction of MR ratio and RA value with annealing at 270 °C for 3 h than the normal junction. From x-ray photoelectron spectroscopy and auger electron spectroscopy depth profiles, it is found that the nitrogen, which was initially at an interface between Al and Fe, diffuses into both the Al and FeMn layers after annealing at 230 °C. It seems that the nitrogen plays an important role in reducing Mn diffusion, as well as in improving the junction properties. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1618381]

Magnetic tunnel junctions (MTJs) have been investigated intensively over the last few years, both for purely scientific reasons and because of their importance in applications such as nonvolatile magnetic random access memory (MRAM) and read heads for high-density magnetic storage. In order to achieve high performance and density in magnetic memory devices, it is desirable for the MTJ to have a high magnetoresistance (MR) ratio and low junction resistance. The junction resistance-area product (RA) is an important factor in determining readout access speed and signal-to-noise ratio in memory applications.[1] The goal that has generally been set for junction resistance for MRAM application is RA<10 k$\Omega\ \mu$m^2 and, for read head application, RA$<10\ \Omega\ \mu$m^2.[2] Although several efforts have been made to provide possible ways of reducing the junction resistance to a few tens or a few $\Omega\ \mu$m^2, the junctions' MR ratio cannot be maintained at the optimal level.[3–5]

In this letter, we present the effects of nitrogen treatment and thermal annealing on MTJs. The nitrogen treatment of a MTJ was carried out by exposing the Fe pinned layer to nitrogen plasma for a few tens of seconds before deposition of an insulating layer. The nitrogen-treated junction shows lower MR ratio and RA values than the untreated normal junction. Remarkably, the nitrogen-treated junction recovers the same MR ratio upon thermal annealing as that of an optimal normal junction, while it retains the low RA value. Moreover, compared with the normal junction, the nitrogen-treated junction shows less reduction of MR and RA after annealing at 270 °C for 3 h. It is found that nitrogen plays an important role in suppressing Mn diffusion as well as in the improvement of junction properties.

The MTJs on Si/SiO$_2$(200 nm) substrate were prepared by using a dc/rf magnetron sputtering system with a base pressure below 8×10^{-8} Torr at room temperature. The structure of the junction consisted of Ta(10)/ NiFe(14)/ FeMn(10)/ NiFe(6)/ Fe(2)/ Al and oxidation/ NiFe(20)/ Au(20), where the numbers in parenthesis are the thickness of each layer in units of nanometers. The thickness of the Al metal layer was 1.32 nm. The tunnel barrier was formed by plasma oxidation for 90 s in an oxygen pressure of 100 mTorr and a dc power of 7 W. The nitrogen treatment was performed at a nitrogen pressure of 100 mTorr by using a dc power of 3.5 W without vacuum breaking for various nitrogen exposure times ($t_{ex}=0$, 10, 30, and 60 s). Detailed descriptions of the fabrication have been described elsewhere.[6] The patterned size of the junctions was $10\times20\ \mu$m^2. Magnetic transport properties of the junctions were measured by using a dc four-probe method at room temperature. The junctions were characterized by x-ray photoelectron spectroscopy (XPS) and auger electron spectroscopy (AES). The XPS measurements were performed with a Mg K_α x-ray source (1253.6 eV) utilizing a modified VG ESCALAB 220 electron energy analyzer. The AES was obtained with an electron beam of 10 keV using a VG MICROLAB 310D.

Figure 1(a) shows the variation of the MR ratio, defined as MR$=(R_{ap}-R_p)/R_p$ where R_{ap} and R_p are the resistances for antiparallel and parallel magnetic configurations between pinned and free layers, as a function of the annealing temperature (T_a) for the nitrogen-treated and normal junctions. The annealing time was 30 min at each temperature. The nitrogen-treated junctions show a much lower MR ratio than

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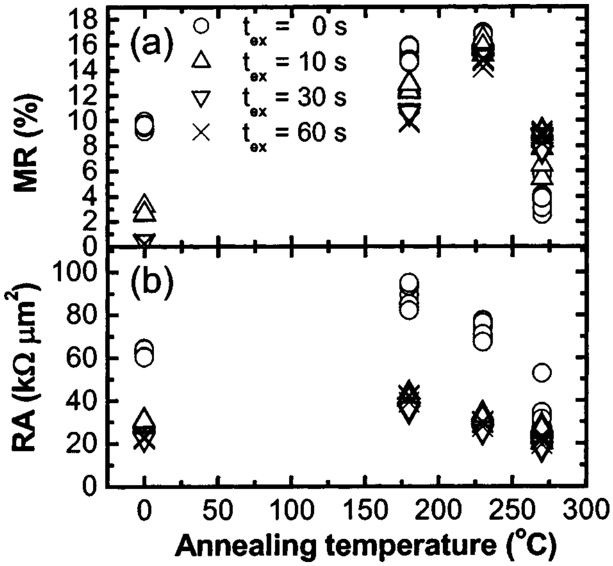

FIG. 1. MR ratio vs annealing temperature for junctions with various nitrogen exposure times (t_{ex}). (b) RA vs annealing temperature.

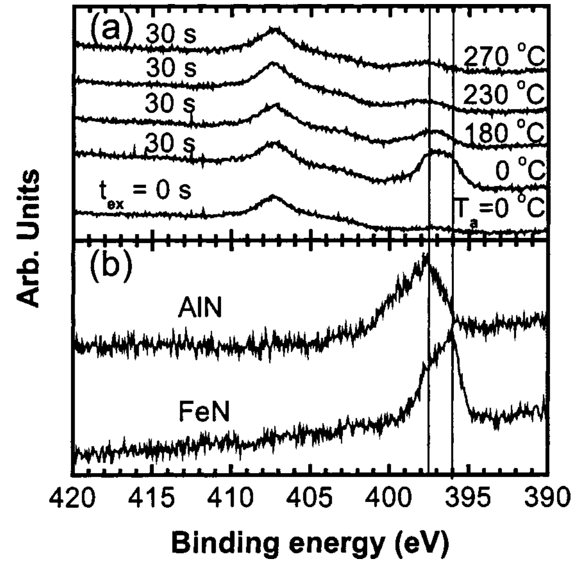

FIG. 2. (a) XPS intensity (N 1*s*) for junctions with nitrogen exposure time of $t_{ex}=0$ and 30 s after thermal annealing at the indicated temperatures. (b) XPS (N 1*s*) intensity in AlN and FeN as a reference. The vertical lines indicate the peaks of 397.5 and 396.0 eV.

the normal junction. The reduction in the MR ratio for the nitrogen-treated junctions depends on t_{ex} and the MR ratio when $t_{ex}=60$ s is about 0%. The MR ratio of a normal junction improves from $\approx 10\%$ to $\approx 17\%$ as T_a increases, up to 230 °C, and then falls at T_a higher than 230 °C. By contrast, the nitrogen-treated junctions show an enhancement of MR ratio with thermal annealing at a much higher rate, resulting in almost the same MR ratios as the optimal value (MR $\approx 17\%$) of the normal junction after annealing at $T_a = 230$ °C. Figure 1(b) shows the variation in RA for the junctions in Fig. 1(a) in terms of T_a. RA values are defined by the resistance at parallel magnetic configuration between the pinned and free layers. The RA of a normal junction is about 60 and 70 kΩ μm^2 before and after annealing at $T_a = 230$ °C, respectively, which is the optimal temperature with maximum MR ratio in Fig. 1(a). Before annealing, the nitrogen-treated junctions show lower RA values than the normal junction and the amount of the RA reduction depends on the t_{ex}. Interestingly, after annealing at 230 °C, the nitrogen-treated junctions still have lower RA values than a normal junction. Even when $t_{ex}=10$ s, the nitrogen-treated junction shows lower RA (≈ 30 kΩ μm^2) at $T_a = 230$ °C than a normal junction (RA≈ 70 kΩ μm^2).

XPS measurement was carried out to investigate the nitrogen in the nitrogen-treated junction without top layers above the Al_2O_3 barrier. The nitrogen 1*s* peaks of XPS for both the nitrogen-treated ($t_{ex}=30$ s) and normal ($t_{ex}=0$ s) junctions are shown in Fig. 2(a) at various T_as and the peaks of FeN and AlN are shown in Fig. 2(b) as a reference. Compared with a normal junction, the nitrogen-treated junction in Fig. 2(a) shows a broad peak of binding energy E_b between 394 eV$\leq E_b \leq$400.9 eV before thermal annealing, indicating that the two binding energies of FeN and AlN overlap. This result means that the nitrogen in the Fe layer infiltrates into the Al layer during the deposition of the Al layer. With increasing T_a, the intensity of the broad peak gradually decreases and the peak position shifts to the higher binding energy. That is, the peak at $E_b = 397.5$ eV can be still observed at $T_a = 230$ °C, whereas the peak at $E_b = 396$ eV almost disappears. Thus, we speculate that the low junction resistance for the nitrogen-treated junction in Fig. 1 is due to the incorporation of nitrogen in the Al layer and that the high MR at $T_a = 230$ °C is probably due to a recovery of spin polarization of the Fe layer during thermal annealing.

Figures 3(a) and 3(b) show the AES depth profiles for Al, N, and Mn in the nitrogen-treated junction with $t_{ex} = 60$ s before and after annealing at $T_a = 230$ °C, respectively. Without thermal annealing, the nitrogen profile overlaps considerably with the Al profile. With thermal annealing at $T_a = 230$ °C, some of the nitrogen moves from the Al layer side to the FeMn layer, as shown in Fig. 3(b). These observations are consistent with the results for XPS measurements, i.e., the reduction of the XPS peak intensity and the shift of the peak position as described earlier.

In order to investigate the process of junction degrada-

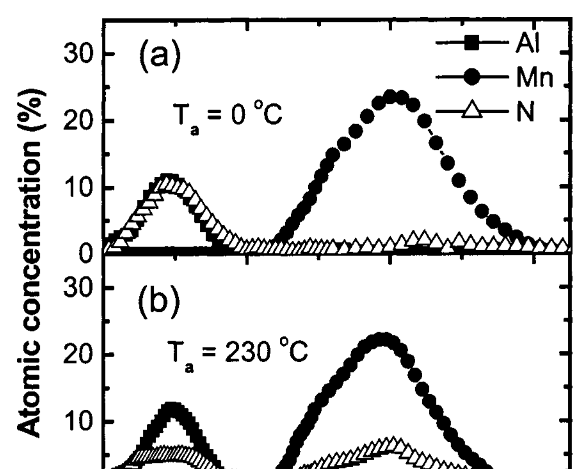

FIG. 3. AES depth profiles for Al, Mn, and N in the junction with nitrogen exposure for $t_{ex}=60$ s before (a) after (b) thermal annealing at $T_a = 230$ °C.

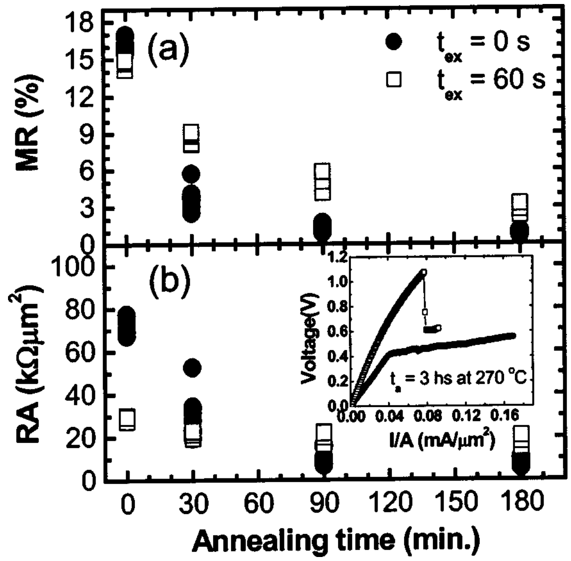

FIG. 4. (a) MR ratio vs annealing time at 270 °C for nitrogen-treated (t_{ex} $=60$ s) and normal junctions, which are optimally annealed at 230 °C. (b) RA vs annealing time. Inset: Current density vs voltage curve for the junctions in (a) after thermal annealing at 270 °C for 180 min.

tion, we carried out thermal annealing at $T_a=270\,°C$ for the optimalized normal and nitrogen-treated junctions with t_{ex} $=60$ s, as shown in Fig. 1. The variations of MR and RA of the junctions are plotted in terms of annealing time (t_a) in Figs. 4(a) and 4(b), respectively. The reduction of MR ratio with an annealing time is clearly lower for the nitrogen-treated junction than the normal junction. While the normal junction shows virtually no MR after thermal annealing for 90 min at $T_a=270\,°C$, the nitrogen-treated junction shows a MR ratio of $\approx 5\%$. For the variation of RA, the nitrogen-treated junction shows significantly smaller reduction, compared with the normal junction. The RA values for the nitrogen-treated junction become larger than those for the normal junction with annealing time of $t_a=90$ min. In addition, we performed the $I-V$ measurement for both annealed junctions for 180 min at 270 °C, which is shown in the inset of Fig. 4(b). The breakdown for a normal junction takes place at about 0.04 mA/μm^2 and 0.42 V and consists of a number of smaller voltage steps. In general, this can be explained by the diffusion of the metal element into the barrier. However, the nitrogen-treated junction reveals the breakdown of a typical insulating barrier at higher current and voltage, resulting in a sudden drop in voltage. In particular, it was reported that the degradation of an annealed junction in MTJ was due mainly to the diffusion of Mn from the antiferromagnetic pinning layer.[7] In this respect, it seems that the diffused nitrogen in the FeMn layer, shown in Fig. 3(b), plays an important role in preventing Mn from diffusing towards a tunnel barrier with thermal annealing.

In conclusion, we observed from XPS and AES data that a small amount of nitrogen, which was initially in-between pinned and insulating layers, diffuses into both insulating (Al_2O_3) and pinning (FeMn) layers with thermal annealing at $T_a=230\,°C$. This nitrogen distribution causes the MTJ to have almost the same MR ratio as, and lower RA than, a normal junction without nitrogen. In addition, the diffused nitrogen in FeMn plays an important role in suppressing the Mn diffusion into the insulating layer with thermal annealing, leading to higher thermal and electrical stability of the nitrogen-treated junction.

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