Change in Co/Al-oxide/Co tunneling junction under constant voltage stress

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Abstract. The tunneling junctions Co(10nm)/Al-oxide/Co(50nm) were fabricated on a glass substrate using a metal mask by ion-beam sputtering. On the bottom Co electrode, a 2.5 nm thick Al film was deposited and was oxidized in pure oxygen gas for 12 - 216 h at 200 C. The electrical properties of the junction were measured by the four-probe method under a constant voltage and the current-voltage characteristics at low voltages were measured every half hour. The barrier parameters of Al-oxide layers are evaluated from a fit to the calculated current-voltage characteristics for the junction by using the Particle Swarm Optimization method with high accuracy. The oxide layer was assumed to comprise two different barriers. In case of the barrier with low potential height, oxidation proceeds by applying the voltage and the resistivity increases. In the case of hard breakdown, the barrier retains uniform at the breakdown with sudden change of the resistivity. In other case of the soft breakdown, the barrier shows segregation just before the breakdown.

1. Introduction
Magnetic tunneling junctions (MTJs), which consist of two ferromagnetic electrodes separated by an insulator, are expected for future memories such as magneto-resistive random memory (MRAM), because of a large magneto-resistance ratio at room temperature [1]. However, MTJs suffer from electrical breakdown by induced voltage stress because of the very thin insulator. It is an important reliability issue for commercial device applications. Various investigations on electrical breakdown of MTJs have been performed [2, 3]. However, the mechanism of the breakdown was not clarified. In this article, the change in the insulator has been studied under constant voltage stress.

2. Experimental and calculation method
The tunneling junctions Co(10nm)/Al-oxide/Co(50nm) were fabricated on a glass substrate using a metal mask by ion-beam sputtering. On the bottom Co electrode, a 2.5 nm thick Al film was deposited and was oxidized in pure oxygen gas for 12 - 216 h at 200 C. The electrical properties of the junction were measured by the four-probe method under a constant voltage more than 0.5 V. And the current-voltage characteristics at low voltages from -0.3 V to 0.3 V were measured every half hour.

Two potential model is used for the evaluation of the barrier parameters. Figure 1 shows the schematic illustration of the model. The insulator film is assumed to comprise two different barriers. Here, the $\phi_a$ and $\phi_b$ are the barrier heights, $d$ is the thickness, and $\beta$ is the ratio of potential barrier thickness. In this model, tunneling probability is numerically calculated from Schrödinger equation. The barrier parameters of Al-oxide layers are evaluated by fitting the current-voltage characteristics.
for the junction to the calculated result obtained from the two-potential model [4].

Figure 2 shows a typical result of the barrier-height dependence of the fitting error, \( Q \). The fitting error is defined as sum of the absolute differences between the observed value and calculated value. In this case, we set parameters \( \phi_a = \phi_b \) and \( d = 1.7 \text{ nm} \), which are obtained as the suitable values by the steepest descend method of a preliminary search. The fitting error oscillates extremely. So the conventional steepest descend method cannot find the real minimum value due to an interruption by local minimum values. In order to solve this problem, the Particle Swarm Optimization method [5] is applied. Table 1 shows that this method can determine the barrier parameters with high accuracy.

| Table 1. Barrier parameters determined by the PSO method with different convergence parameters. |
|---|---|---|---|
| \( d \) (nm) | \( \phi \) (eV) | \( Q \) | endstep |
| Steepest descent method | 1.80 | 0.90 | 2.78 | 1200 |
| PSO method (1) | 1.67 | 1.07 | 1.41 | 617 |
| (2) | 1.66 | 1.09 | 1.41 | 728 |
| (3) | 1.69 | 1.04 | 1.42 | 482 |

3. Results and discussion

3.1. Progress of oxidation in the Al-oxide layer

Figure 3 shows the change in tunnel resistance under constant voltage stress at 0.5 V. The change in thickness and barrier heights evaluated are shown in Figures. 4(a) and (b).
At first, the oxide layer is almost the homogeneous insulator ($\beta \sim 0$) with low barrier height of about 0.64 eV, which is estimated to be AlO$_2$ by XPS analysis. Then oxidation proceeds by applying the voltage and the resistivity increases. The phase changes to Al$_2$O$_3$ with high barrier height of about 0.72 eV at 15 h. The total thickness of about 2.3 nm does not change during the oxidation process, so the density of the oxide layer increases. Finally, the oxide layer becomes a homogeneous ($\beta \sim 1$) and dense Al$_2$O$_3$ phase with high barrier height.

Figure 5 shows the change in tunnel resistance at voltage of 0.6 V. The change in thickness and barrier heights evaluated are shown in Figures. 6(a) and (b). The parameters hardly change at 0.6 V. The Al-Ox phase is almost the homogeneous, because $\phi_a \sim \phi_b$.

Figure 4. Change in thickness and barrier heights evaluated under constant voltage stress at 0.5 V.

Figure 5. Change in tunnel resistance at voltage of 0.6 V.

Figure 6. Change in thickness and barrier heights evaluated at voltage of 0.6 V.
3.2. Hard breakdown of the Al-oxide layer
As shown in Figure 7, changes in tunnel resistance, evaluated thickness and barrier heights at voltage of 0.65 V are almost the same as changes at 0.6 V. However, a sudden decrease in the tunnel resistivity, which means an electrical breakdown, occurs at about 39 h. The barrier parameters do not change before the breakdown. It is thought that the breakdown is probabilistic.

![Figure 7. Change in tunnel resistance at voltage of 0.65 V.](image)

3.3. Soft breakdown of the Al-oxide layer
Figure 8 shows the change in tunnel resistance at voltage of 0.6 V for another specimen. The change in thickness and barrier heights evaluated are shown in Figures 9(a) and (b). There are sudden changes in tunnel resistance and barrier parameters. These changes, called as soft breakdown, seem to be caused by a reaction such as phase change in the insulator.

![Figure 8. Change in tunnel resistance at voltage of 0.6 V for another specimen.](image)

![Figure 9. Change in thickness and barrier heights evaluated at voltage of 0.6 V.](image)
4. Conclusions
There are several different cases of the change under constant voltage stress. In case of the barrier with low potential barrier, oxidation proceeds by applying the voltage and the resistivity increases. The barrier retains uniform at the breakdown with abrupt change of the resistivity. However, the barrier sometimes shows segregation, which leads to a soft breakdown.

References
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