# Nearly ideal spin tunneling efficiency by lowering the trap density at an amorphous-MgO / $n^+$ -Si(001) interface with a SiO<sub>x</sub> insertion layer

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### Abstract

We achieved nearly ideal spin tunneling efficiency  $\eta$ by lowering the interface trap density  $D_{it}$  with a SiO<sub>x</sub> layer in Fe / Mg / amorphous-MgO(1.0 - 1.5 nm) / plasma-oxidized SiO<sub>x</sub>(~0.2 nm) /  $n^+$ -Si(001) junctions. We estimated the tunneling electron spin polarization  $P_S$  in Si by narrower three-terminal Hanle signals measured at 4 K. At the optimum MgO thickness and oxidation time,  $P_S = 41\%$  which is nearly equal to the spin polarization  $P_{FM}$  (= 44%) of Fe at the Fermi level, and  $\eta = P_S/P_{FM} =$ 0.93. By quantitatively estimating  $D_{it}$  and  $P_S$  of various junctions, we show that lowering  $D_{it}$  is crucial to obtain  $\eta \approx 1$ .

#### 1. Introduction

To realize Si-based spin-functional devices with a high magneto-current ratio, spin injection/extraction into/from Si through a ferromagnetic metal(FM)/MgO/Si tunnel junction has been extensively studied [1-3]. For such devices, highly-efficient spin injection/extraction is required and it can be measured by spin tunneling efficiency  $\eta = P_{\rm S}/P_{\rm FM}$ , where  $P_{\rm S}$  is tunneling electron spin polarization in Si and  $P_{\rm FM}$  is the spin polarization of FM at the Fermi level. Although  $\eta = 1$  is ideal, the  $\eta$  values reported so far were significantly smaller than 1;  $\eta = 0.17$  at 8 – 300 K in a Fe/MgO/ $n^+$ -Si junction [1] and  $\eta = 0.41$  at 300 K in a Co<sub>2</sub>FeSi/MgO/Si junction [2]. In our previous paper, we found that the spin polarization of electrons passing through a ferromagnetic tunnel junction is reduced by a magnetically-dead layer formed at the FM/MgO interface, and demonstrated that an ultra-thin Mg insertion layer effectively suppresses the magnetic degradation [3]. In such junctions, we obtained  $\eta = 0.41$  at 4 K, but it is not high enough. On the other hand, since electron trap states at the MgO/Si interface can be spin flip centers,  $\eta$  is possibly improved by lowering the interface trap density  $D_{it}$ , although it has not been demonstrated ever. In this study, we show that nearly ideal  $\eta = 0.93$  is achieved in a Fe/Mg/MgO/Si junction by lowering D<sub>it</sub> with a plasma oxidation of the MgO/Si structure during the junction formation.

#### 2. Device preparation

The sample preparation was as follows: After thermal cleaning of a H-terminated Si(001) substrate in an ultra-high

vacuum, a MgO layer with the thickness  $d_{MgO} = 1.0, 1.2,$ and 1.5 nm was deposited at room temperature by electron beam (EB) evaporation. Subsequently, the MgO/Si structure was oxidized at room temperature by a RF plasma source for oxidation time  $t_{ox} = 1$  and 3 min, during which an ultrathin  $SiO_x$  layer was formed between MgO and Si. A non-oxidized MgO/Si structure was also prepared, and it is denoted by  $t_{ox} = 0$  min. On this surface, three types of structure were prepared at room temperature: (from top to bottom) (1)For three-terminal Hanle measurements, an Al(10 nm)/Mg (1 nm)/Fe (3 nm)/Mg (1 nm) was deposited on a MgO( $d_{MgO} = 1.0, 1.2, \text{ and } 1.5 \text{ nm}$ ) layer by molecular beam epitaxy (MBE), (2)For C-V measurements and the conductance method with MOS capacitors[4], an Al(10 nm)/MgO (4 nm) was deposited on a MgO( $d_{MgO} = 1.2$  nm) layer by EB evaporation and MBE, and (3)For X-ray photoelectron spectroscopy (XPS) measurements, a 3-nm-thick Al was deposited on a MgO( $d_{MgO} = 1.0$  nm) layer by MBE. After exposure to air, the fabrication process of the top and bottom electrodes for (1) and (2) was the same as that in the previous paper [3]. Phosphorus-doped Si(001) substrates with ~1 m $\Omega$ cm and ~100 m $\Omega$ cm were used for (1)(3) and (2), respectively. The diameter of a circular pillar junction for (1) was 17.8 μm, whereas that for (2) was 178 μm. Figure 1 shows a three-terminal device structure with the junction structure (1) and the three-terminal Hanle measurement setup, in which the voltage change  $\Delta V^{N}$  is measured with a constant negative current  $I_{\rm B}$  (spin extraction geometry) driven from the top to the bottom electrodes while a perpendicular magnetic field is swept from -3 to 3 kOe.



Fig. 1 Schematic structure of a device with multi-layered magnetic junctions and the three-terminal Hanle measurement set up.

#### **3.** Characterization of SiO<sub>x</sub> formation

From XPS spectra of Si 2*p* in Fig. 2(a), the sample with  $d_{MgO} = 1.0$  nm and  $t_{ox} = 0$  min does not have SiO<sub>x</sub>, whereas

the sample with  $d_{MgO} = 1.0$  nm and  $t_{ox} = 1$  min has a SiO<sub>x</sub> layer with  $x \sim 2$ . By the angle-resolved XPS spectrum of Si 2*p*, the thickness of SiO<sub>x</sub>  $d_{SiOx}$  was estimated to be 0.26 nm. Figure 2(b) shows resistance-area product *RA* at zero bias of all the three-terminal samples, which were estimated from the *I-V* curves measured at 4 K. For the same  $d_{MgO}$ , *RA* increases exponentially with increasing  $t_{ox}$ , indicating that  $d_{SiOx}$  increases with increasing  $t_{ox}$ .



Fig. 2 (a) X-ray photoelectron spectroscopy (XPS) spectra of Si 2*p* measured, where black and blue curves are the spectra of the Al(3 nm)/MgO(1 nm)/Si ( $t_{ox} = 0$  min) and Al(3 nm)/MgO(1 nm)/SiO<sub>x</sub>/Si ( $t_{ox} = 1$  min), respectively. (b) Resistance-area product (*RA*) at zero bias voltage of the junction plotted as a function of  $t_{ox}$ , which were estimated from *I-V* characteristics measured at 4 K. The open black circles, open green squares, and open orange triangles are the values for MgO thickness  $d_{MgO} = 1.0, 1.2, \text{ and } 1.5$  nm, respectively.

#### 4. Spin extraction experiments and estimation of $\eta$

Figure 3(a) shows Hanle signals  $\Delta V^{N}$  of the devices with  $d_{MgO} = 1.2$  nm measured at 4 K with  $I_{B} = -30$  mA, where black, blue, and red curves are  $t_{ox}= 0$ , 1, and 3 min, respectively. We estimated the spin lifetime  $\tau_{S}$  and  $P_{S}$  by fitting a theoretical curve [3], which is the light green curve superimposed on each signal. Whereas  $\tau_{S}$  was almost the same among the devices,  $P_{S}$  was changed depending on  $t_{ox}$ . In the same manner,  $P_{S}$  in all the devices were estimated, and these are plotted as a function of  $t_{ox}$  in Fig. 3(b). Interestingly, as  $t_{ox}$  increases,  $P_{S}$  (and  $\eta$ ) for the devices with  $d_{MgO} = 1.2$  and 1.5 nm increase at first, show the maximum at  $t_{ox} = 1$  min, and then slightly decrease. This behavior



Fig. 3 (a) Narrower three-terminal Hanle signals  $\Delta V^{\rm N}$  of devices with  $d_{\rm MgO} = 1.2$  nm measured at 4 K with  $I_{\rm B} = -30$  mA, where black, blue, and red curves are  $\Delta V^{\rm N}$  of the devices with  $t_{\rm ox} = 0, 1$ , and 3 min, respectively, and the light green curve superimposed on each  $\Delta V^{\rm N}$  is the fitting curve with Eq. (2) in [3]. (b) Tunneling spin polarization  $P_{\rm S}$  in Si and spin tunneling efficiency  $\eta$  plotted as a function of  $t_{\rm ox}$ , where each value was estimated from  $\Delta V^{\rm N}$ measured at 4 K with  $I_{\rm B} = -30$  mA. The open black circles, open green squares, and open orange triangles are the values for  $d_{\rm MgO} =$ 1.0, 1.2, and 1.5 nm, respectively.

cannot be explained by the  $RA - t_{ox}$  relation in Fig. 2(b), in which *RA* increases with increasing  $t_{ox}$ .

We also measured the  $I_{\rm B}$  dependence of  $P_{\rm S}$  of the device with  $d_{\rm MgO} = 1.2$  nm and  $t_{\rm ox} = 1$ min, and found that  $P_{\rm S}$  increases with decreasing  $I_{\rm B}$ . At  $I_{\rm B} = -15$  mA, we obtained the maximum value of  $P_{\rm S} = 0.41$ , which corresponds to  $\eta = 0.93$ .

#### 5. Relation between Ps and interface trap density Dit

Figure 4 shows  $D_{it}$  estimated from the conductance method at room temperature, where  $d_{MgO} = 1.2$  nm and black, blue, and red circles are  $t_{ox} = 0$ , 1, and 3 min, respectively. As  $t_{ox}$  increases from  $t_{ox} = 0$  min,  $D_{it}$  in whole the energy range decreases at  $t_{ox} = 1$  min, and then it slightly increases at  $t_{ox} = 3$  min. This behavior is highly correlated with the  $t_{ox}$  dependence of  $P_S$  for  $d_{MgO} = 1.2$  nm in Fig. 3(b); the device with lower  $D_{it}$  shows higher  $P_S$ . Thus, we concluded that the highest  $P_S = 41\%$  ( $\eta = 0.93$ ) in this study is achieved by lowering  $D_{it}$  at the insulator/Si interface.



Fig. 4 Interface trap density  $D_{it}$  in capacitors with  $d_{MgO} = 1.2$  nm and  $t_{ox} = 0, 1$ , and 3 min plotted by electron energy E, where the origin of E is set at the conduction band edge  $E_c$  of Si, the middle of the bandgap of Si is indicated by "Mid gap", and  $E_F$  denotes the Fermi level of the Si substrate. The black, blue, and red circles are  $D_{it}$  in the capacitors with  $t_{ox} = 0, 1$ , and 3 min, respectively.

## 6. Conclusion

We have achieved the spin tunneling efficiency  $\eta = P_{\rm S}/P_{\rm FM}$  value of 0.93, which is the highest ever reported by lowering  $D_{\rm it}$  at the insulator/Si interface with the SiO<sub>x</sub> (x ~2) insertion layer. The  $\eta$  obtained in this study is nearly ideal in the junctions using an amorphous insulator layer, *i.e.*, without the spin filter effect.

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