Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2015

## Supporting Information for

# Effects of radio-frequency sputtering power of MgO tunneling barrier on tunneling magneto-resistance ratio for Co<sub>2</sub>Fe<sub>6</sub>B<sub>2</sub>/MgO-based perpendicularmagnetic tunnel junctions

Du-Yeong Lee, Hyong-Tak Seo and Jea-Gun Park\*

<sup>†</sup>Department of Electronics and Computer Engineering, Hanyang University, Seoul, 133–791, Republic of Korea E-mail: parkjgL@hanyang.ac.kr



**Supplement 1.** Effect of *in-situ* annealing on the TMR ratio enhancement. (a) without annealing (b) *in-situ* annealing at 400°C for 30 min after b.c.c capping-layer sputtering, and (c) *in-situ* annealing at 400°C for 30 min after MgO barrier sputtering.



**Supplement 2.** Effect of *ex-situ* annealing on the *M-H* curves for  $Co_2Fe_6B_2$  free layer on Ta seed. In general, the  $M_s$  (saturation magnetization) of 1.05-nm thick  $Co_2Fe_6B_2$  free layer sputtered on Ta seed is about 70~80 µmeu. We could obtain ~90 µemu of 1.05-nm thick  $Co_2Fe_6B_2$  free layer sputtered on b.c.c seed, as shown in Figs. 2(b)-(e).



**Supplement 3.** Dependency of the crystallinity of a thick MgO layer (~10 nm) on the MgO sputtering: (a) 250, (b) 300, (c) 400, and (d) 500 W. The detailed vertically stacked samples were made of a 12-inch Si wafer /SiO<sub>2</sub> layer (100 nm) /Ta seed (5 nm)  $/Co_2Fe_6B_2$  (3 nm) /MgO tunneling barrier (10 nm, RF power: 250, 300, 400 and 500 W)  $/Co_2Fe_6B_2$  (3 nm) /Ta capping layer (2 nm) layer. All samples were subject to three times annealing at 350°C for 1 hr and 400°C for 30 min and followed by crosssectional TEM observation. Overall MgO layers for all samples showed well crystallized except at  $\sim 1$  nm of the interface between the MgO layer and Co<sub>2</sub>Fe<sub>6</sub>B<sub>2</sub> layer, which is consistent with the growth mechanism of a thick MgO layer explained by the reference [J. Phys. D: Appl. Phys., 40 (2007) R337–R354.]. However, from 500 to 300 W, the roughness (i.e., thickness variation) of the thick MgO layers decreased with the RF sputtering power of the thick MgO layers since the compositional roughening of the MgO layer decreased with the RF sputtering power of the MgO layer, as shown in Supplement 4(a)-(c). This result enables us to prospect that the thickness variation of thin (~1.15 nm) MgO layers also would decrease with the RF power of thin MgO layers, as compared by Figs. 3(a)-(c) and supplement 4(a)-(c). In particular, a severe compositional roughening of a thin (~1.15 nm) MgO layer at a high RF sputtering power such as 500 W would cause a local amorphous region a thin (~1.15 nm) MgO layer, as shown in Fig.3(a) at our manuscript. Otherwise, for the thick (~10 nm) MgO layer sputtered at 250 W, the MgO layer at ~1 nm of the interface between the MgO layer and CoFeB layer showed a locally amorphous layer mixed with a crystallized layer, as shown Supplement 3(d), well correlated with Fig. 3(d) in our manuscript. Thus, this result enables us to prospect that a thin (~1.15 nm) MgO layer could be an almost amorphous layer, compared by Fig. 3(d) and Supplement 3(d). In addition, This result would be associated with that too low RF sputtering power, i.e., less than 250 W, would produce an almost amorphous MgO layer since the energy to achieve the complete crystallization of the MgO layer would not be sufficient at 250 W.



**Supplement 4.** Dependency of XRD peak distribution on RF sputtering power for 10nm MgO tunneling barrier *ex-situ* annealed at 400°C. The MgO (200) peak was ~0.5° shifted from 44.2° to 43.7° while the Si (100) peak did not change, which was probably related to too thin MgO thickness (~10 nm) or XRD calibration issue. However, it was confirmed that the MgO of ~10 nm showed the MgO (200) crystallinity in an x-TEM image in Fig. 4 (b) at our manuscript.



Supplement 5. Dependency of roughness and density of ~10 nm-thick MgO layer on RF sputtering power measured by x-ray reflection (XRR) measurement: 250 W and 300 W. The roughness of the MgO layer (~10 nm) sputtered at 250 W (~2.11 nm) was lower than that at 300 W (~2.47 nm). This result is probably attributed to that the MgO layer sputtered at 250 W includes ~1.0 nm of almost amorphous layer close to the interface between the MgO layer and Si substrate while the MgO layer sputtered at 300 W is well crystallized at the interface, as shown in x-TEM images in Fig. 4(b) in our manuscript. Note that the interface roughness for amorphous thin film is less than that for crystallized thin film. In addition, the density of the MgO layer (~10 nm) sputtered at 250 W (5.11 g/cm<sup>3</sup>) was higher than that at 300 W (3.50 g/cm<sup>3</sup>). In general, the density of the crystallized MgO layer is 3.58 g/cm<sup>3</sup>, which is a similar to the density of the MgO layer sputtered at 300 W. However, the density of the MgO layer sputtered at 250 W was very high since the MgO layer sputtered at 250 W includes W includes ~1.0 nm of almost amorphous layer close to the interface between the MgO layer and Si substrate. Thus, the XRR results imply that the roughness difference of the MgO layer sputtered between 250 and 300 W would not be related to the TMR ratio difference of the MgO layer sputtered between 250 and 300 W. But, the density difference of the MgO layer sputtered between 250 and 300 W would be rather related to the TMR ratio difference of the MgO layer sputtered between 250 and 300 W.