Annealing Effects On Sequentially Sputtered Co - HfO_2 Granular Films.

M. Chadha, V. Ng*
Information storage Materials laboratory, National University of Singapore, Singapore 117576

Granular films comprise of well-defined nanometer size magnetic particles embedded in an insulating matrix. Conduction between magnetic grains by electron tunneling through the insulator barriers gives rise to the tunneling magneto resistance (TMR) phenomenon [1]. Previous works on Co-Al_2O_3 granular systems showed a high content of cobalt oxide. The antiferromagnetic nature of cobalt oxide decreases TMR effect and is thus not desirable in the film[2-3]. In the present work we investigate the effect of hafnium oxide used as insulating matrix in cobalt magnetic film to decrease the possibility of formation of cobalt oxide during sputtering. Our motivation is related to the heat of formation of hafnium oxide being significantly higher than other oxides [4]. Annealing was also employed to encourage the Hf-rich film to replace any remaining cobalt oxide in the film, thus forming more Co in the HfO_2 matrix.

Discontinuous Co and HfO_2 layers were sequentially sputtered on glass and Si substrates using argon at 5 mTorr and a base pressure of ~10^{-8} Torr, giving total thickness of 20 nm. Post deposition thermal annealing was carried out in vacuum at ~10^{-7} Torr. The films were annealed for 20 min at increasing annealing temperatures (T_A) from 323K to 475K in steps of 50K. Magnetoresistance (MR) of the films was measured at room temperature by DC four-point probe setup. We measured R_0 the resistance value at zero field and ΔR (difference between R_0 and resistance at 4 k Gauss maximum field). The magnetic properties were measured using the vibrating sample magnetometer (VSM). X ray photoelectron spectroscopy (XPS) with an AlKα radiation source was also performed.

Figure 1 presents the XPS spectra for as-sputtered and annealed samples, showing a Co $2p_{3/2}$ peak at the electron binding energy of 778.3 eV. A small cobalt oxide shoulder at 782.5 eV, seen more clearly in the zoomed in picture, refers to various cobalt oxides present in the as-sputtered sample. This amount is generally smaller than that present in other Co-Al oxide granular films [2]. Since HfO_2 is more thermodynamically favorable than Cobalt oxide. We postulate that HfO2 has replaced Co in Cobalt oxide for the annealed films as the cobalt oxide peak was smoothened out with annealing, shown representatively at 473 K in the zoomed in picture. Figure 2 shows R_0, ΔR and coercivity (Hc) vs. T_A. These trends agree well with XPS results. The increasing trend for R_0 values is attributed to the presence of more HfO_2. The saturation magnetization, obtained from VSM analysis also showed an increasing trend supporting the increase in pure cobalt content. This decrease in the antiferromagnetic cobalt oxide causes a decrease in the current component independent of spin polarization, hence increasing ΔR from 272Ω for as sputtered to 1980Ω for the sample annealed at 373K. The MR ratio defined as $\Delta R/R_0$ also showed an increase from 1.1% to 3.0% from the as-sputtered film to that annealed at 373K (inset of Fig 2).

Annealing at temperatures above 373K reduces structural disorder and increases the electron free path, explaining the decrease in R_0. Due to an increase in the Co content we postulate that there is an increase in the grain size as previously also shown by Gang Xiao[5]. This has an effect on decreasing the magnetic scattering on the surface and thereby decreasing ΔR and thus the MR ratio.

In conclusion we demonstrated that HfO_2 is effective in producing granular film having low cobalt oxide content and a substantial MR ratio. In addition, annealing further replaced cobalt oxide with HfO_2 and hence enhancing the TMR ratio. We will present the Transmission electron microscopy analysis of the annealing effects on the film microstructure of Co-HfO_2 granular system.

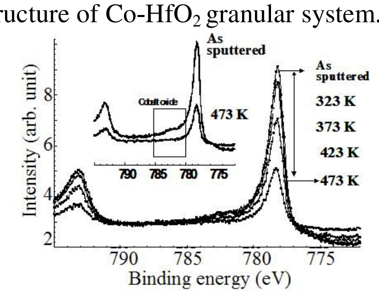

Fig 1. Scans of Co 2p for as sputtered and samples annealed at 323K to 473K.Instead show the Cobalt Oxide peak for as sputtered & 473K sample (Y axis intestinally modified)

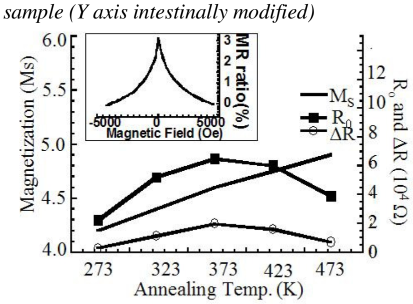

Fig 2. Trends in R_O, ΔR and Hc vs.T_A. Instead shows MR ratio for sample annealed at 373K.

[1] M.Ohnuma, et al, JAP, 82, pp.5646,1997.[2] K.Asami, et al, Surf. Interface Anal.,28, pp.250, 1999.[3] I.Y. Hwang , et al, Surf. Interface Anal.,35, pp.250 [4] Handbook of Materials Sc, VolI (1974).).[5] Jian-Qing Wang, et al, Phy rev.,49, pp.3982, 1994

* Email: elengv@nus.edu.sg