TEM Investigation of MgO Thin Films for Magnetic Tunnel Junction Application

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Abstract. TEM analysis of sputter-deposited MgO has been performed using a plan view and cross-section approach. The influence of the sputtering parameters including target to substrate distance (TSD), sputtering power and Ar pressure on the structure and composition of deposited films has been investigated by analysing in plan view. Optimum deposition conditions have been identified and MgO layers deposited under these are partly crystalline within an amorphous matrix. Cross-sectional TEM imaging of MgO deposited under optimum conditions onto CoFeB in a basic MTJ structure (CoFeB/MgO/CoFeB trilayer), shows that in the initial deposition, at the bottom interface of the trilayer, amorphous MgO forms and this then becomes partly crystalline (but with no preferred texture) with continued deposition, towards the top interface of the layer. TEM- energy dispersive X-ray (EDX) and electron energy loss spectroscopy (EELS) composition analyses suggest that the inhomogeneous oxide film structure could be caused by the deposition of disordered, non-stoichiometric MgO.

1. Introduction
CoFeB/MgO based magnetic tunnel junctions have attracted increasing attention due to the demonstration of large tunnelling magnetoresistances (TMR) which are promising for non-volatile and high density data storage device applications. The key point is the development of a CoFe(100)\{110\}/MgO(100)\{100\}/CoFe(100)\{110\} orientation in the as-annealed structures because crystalline MgO lattice matching of CoFeB at the interfaces acts as a spin filter allowing only particular electron waves to coherently tunnel across the junction. TMRs of over a thousand percent have been theoretically predicted for such coherent lattice matching structures [1-3]. Nevertheless, TMRs of only hundreds of percent have been reported so far in devices prepared by sputter-deposition, with the shortfall being attributed to the fabrication of non ideal structures.

In order to improve fabrication, the crystallization and growth mechanisms of the active trilayer and the relationship to the sputtering parameters should be investigated. Here we present a fundamental study of thin films sputter-deposited on carbon support films commonly used for TEM (using the combination of dc-rf magnetron sputtering). Plan view TEM imaging, selected area electron diffraction (SAED) and energy dispersive X-ray (EDX) spectroscopy, and electron energy loss spectroscopy (EELS) were employed to study the resulting nanostructure of the films both as-deposited and post-annealing. The MgO sputtering parameters such as target substrate distance (TSD),
sputtering power and Ar gas pressure have been optimized to produce the best crystallinity and microtexture of the deposited films. A cross-section of an active trilayer prepared under these optimum deposition conditions has also been analysed by TEM.

2. Experiment

The samples were produced by using dc-rf magnetron sputtering with a base pressure of $10^{-7}$ mTorr. MgO thin films were sputter-deposited using a sintered MgO target with 99.99 % purity, 50 mm diameter, supervac grade obtained from the Testbourne Company. The effect of sputtering parameters including target to substrate distance (TSD), Ar gas pressure and sputtering power were examined. The TSD was originally at 15 cm, and was then adjusted to 11 cm. Ar gas pressures were 1, 5 and 10 mTorr. Sputtering gun powers were 75, 125 and 175 watts. The plan view samples were prepared by directly depositing on continuous carbon film TEM support grids (Agar Scientific Ltd.). A multilayer sheet structure of a simple MTJ was also produced for cross-sectional TEM analysis. The stack sequence of this MTJ is Ta[7]/CoFeB[4]/MgO[12]/CoFeB[8]/Ta[5], the numbers in parenthesis are estimated thickness in nanometers. The cross-sections were prepared by conventional TEM sample preparation using ion milling. Nanostructural analysis was performed using a Philips CM200 field emission gun TEM fitted with an Oxford Instruments ultrathin window ISIS energy dispersive X-ray (EDX) analyser and a Gatan Imaging Filter (GIF 200). Composition was monitored in the TEM with spot-EDX (the Mg/C and Mg/O ratios were determined from the ISIS processing software using virtual standards for the C, O and Mg Kα X-ray peaks) and EELS. Bulk energy loss spectra were taken in diffraction mode (image coupled) with an approximately 0.18 µm diameter selected area aperture (SAD) inserted, leading to a collection semi-angle of 6 mrad, and a convergence semi-angle of approximately 1 mrad. EEL spectra were acquired with an energy dispersion of 0.3 eV/channel and an energy resolution of 0.8eV.

3. Result

3.1. Target to substrate distance (TSD) effect

TEM images of the MgO films deposited on carbon film supports at the two TSDs are shown in figure 1. Both films were sputtered at 175 watts and 1 mTorr Ar pressure and both films have MgO crystallites approximately 8-10 nm in size embedded in an amorphous matrix. Denser clusters of agglomerated crystallites can be observed at the 11 cm TSD. The EDX Mg/C ratio increases at the shorter TSD suggesting a higher deposition rate at 11 cm TSD. Both films were deposited for the same length of time and yet despite the increased deposition rate (and therefore thickness of the film) at 11 cm TSD, a continuously crystalline MgO layer has not been achieved.

The differences in film crystallinity could be because at the shorter TSD the deposited species have, high surface mobility and agglomerate, as seen in figure 1 e) and f). When a substrate is further away from a sputtering target the sputtered atoms experience a higher number of collisions before approaching the substrate and therefore on average have lower kinetic energy on deposition. The EELS oxygen K-edges in the two samples are also compared in figure 2, the edge of the 15 cm film exhibits less fine structure than that of the 11 cm film and it has been reported that this loss of the EELS extended fine structure is indicative of disorder or fewer surrounding oxygen atoms [4]. A shoulder just above the edge onset is also present in the 15 cm film (marked by the arrow) but not the 11 cm one. This feature is believed to be due to gap states that reflect band tailing in the conduction band and has been attributed to vacancies or other structural defects in a MgO/Fe structure [4-5]. The oxygen K-edge of the 11 cm film matches very closely that of bulk MgO. Therefore we suggest that there is a significant concentration of defects in the film deposited at 15 cm TSD.
3.2. Sputtering power and Ar pressure effect

The EDX Mg/C ratio increases with increasing sputtering power and decreases with increasing Ar pressure (figure 3 and 4 respectively). Assuming the carbon films to be of approximately constant thickness suggests that the deposition rate is directly proportional to the sputtering power but inversely proportional to the Ar pressure. At a TSD of 11 cm, 1 mTorr Ar pressure and 75 watts a very thin amorphous MgO film was deposited (not shown here). Very few numbers of crystallites form when sputtering at 125 watts, and much larger numbers are observed at 175 watts (as shown in figure 1b). Varying the Ar gas pressure at 175 watts from 1 to 5 mTorr does not significantly change the film structure apart from producing slightly less crystallite agglomeration at 5 mtorr. Only isolated crystallites are observed at 15 mTorr Ar Pressure. EDX Mg/O values for all of the films are non-stoichiometric, but approach stoichiometry in the films grown at high power and low Ar pressure.

We believe that the relative amount of disordered/amorphous material in the deposited MgO films is consistent with the change in Mg/O ratio, suggesting that the amorphous phase is non-stoichiometric.
3.3. Cross-sectional TEM

Cross-sectional TEM images of the MTJ active trilayer fabricated using the optimum sputtering conditions determined above show an MgO layer, approximately 10 nm thick (figures 5). The MgO barrier was made deliberately thicker than a working MTJ device to be comparable to the MgO films produced for plan view analysis. This MgO barrier is amorphous in the first few nanometres above the bottom CoFeB electrode (initial stage of deposition): no lattice fringes were visible despite tilting ±10° away from the interface plane. Crystalline MgO grains are then evident further up the layer increasing in number towards the top (final stage of MgO deposition). The oxygen intensity profile determined using EELS (white line in figure 5b) shows an oxygen rich content in the first few nanometres of MgO deposition. A textured, crystalline MgO layer should develop immediately upon deposition on the smooth surface of the amorphous electrode. Clearly this is not happening and may be a result of disorder due to an initially oxygen rich deposition or surface roughening of the bottom electrode by O ions released by the sputtering of the MgO [4]. Finally, introducing a thin Mg layer prior to the MgO deposition may compensate for the initial oxygen-rich deposition and prevent the oxidative roughening of the bottom electrode [4].

![Cross-sectional TEM image](image)

**Figure 5.** TEM cross-section of the MTJ prepared by the optimum sputtering conditions. a) low magnification image showing a rougher MgO/CoFeB top interface than the bottom one. b) a magnified inset of a), MgO crystallites are seen towards the top of the junction whilst the first few nanometers at the bottom interface are amorphous or disordered, the white line on the left shows the oxygen K-edge intensity which indicates an initial O-rich deposition, producing disordered MgO. The black line is the intensity profile of the associated scanning(S)TEM-high angle annular dark field (HAADF) image that indicates approximately uniform TEM-specimen thickness within the barrier. Measured lattice spacings in regions A, B and C suggest, A = MgO (200) B = MgO (200) or CoFe(110), and C = MgO(111), d_{MgO(200)} = 2.1 Å, d_{CoFeB(110)} = 2.0 Å and d_{MgO(111)} = 2.4 Å.

4. Conclusion

This work has demonstrated the sensitivity of sputtering parameters on the structure and composition of sputter-deposited MgO barriers, destined for magnetic tunnel junction application. All of the deposited MgO layers consist of MgO crystallites in an amorphous matrix. Films with a greater fraction of amorphous material exhibit poorer MgO stoichiometry. Shortening the target to substrate distance, increasing the sputtering power and lowering the Ar gas pressure produce films that have more crystallinity, less defects, improved texture and increased MgO stoichiometry.

**References**

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