# Growth of Magnesium Oxide Thin Films Using Molecular Beam Epitaxy

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**Abstract:** Ultra thin MgO films (1-2 nm) are grown on $Si/SiO_2$ substrates using molecular beam epitaxy (MBE). Different growth conditions such as substrate temperatures, annealing conditions and growth rates are tested in order to achieve optimum parameters for the smooth and uniform growth. The films were characterized ex-situ using atomic force microscopy and x ray diffractio. Our results demonstrate MBE growth of MgO films with an rms roughness better than 0.5 nm on $Si/SiO_2$ substrates. These results are important for the applications of MgO films as tunnel barriers in spintronic devices.

**Keywords:** Ultra thin films, MgO, molecular beam epitaxy (MBE), atomic force microscopy (AFM), X-ray diffraction (XRD).

## INTRODUCTION

Metal oxide thin films are the most important materials because for wide range of applications in electronic, optoelectronic and spintronic devices [1-3]. In particular, the use of MgO tunnel barriers in spin-valve devices has increased because of the fact that these barriers ensure effetive spin injection, high tunnel magnetoresisrance (TMR) and unprecedented thermal stablity [3-5].

For any spin-valve device the most important aspect is the high quality atomically smooth ultra thin tunnel barriers. Thickness fluctuations can create pinholes or short in regions where the barrier is too thin or make the barrier insulating in regions of large thickness. The barrier must have minimum surface roughness as large roughness of the barrier can lead to inhomogeneous currents through the layer that can results local heating and breakdown of the barrier. Among various growth techniques used for the growing thin films, molecular beam epitaxy (MBE) is the most versatile and reliable one because it is capable of depositing high quality layers with abrupt interfaces [6].

In this work, MgO thin films are grown on $Si/SiO_2$ substrates by MBE under different growth conditions such as substrate temperature and annealing conditions [7]. The main task is to reduce the rms roughness of MgO films to a level that make these films suitable for applications as tunnel barriers in spintroncic devices. We have achieved the growth of high quality films with rms roughness better than 0.5 nm without involving any additional surface treatments [8].

## EXPERIMENTAL

The MgO films were grown by MBE by using E-beam evaporation from the MgO source material placed in a graphite crucible. The deposition rate for MgO was kept at 0.007 nm/s. The base pressure in the MBE camber was $2 \times 10^{-10}$ mbar and the substrates were introduced into the chamber via a load lock. The structural quality of the films is studied by x ray diffraction (XRD). The surface morphology of the as grown films is examined ex-situ by an atomic force microscope (AFM) (Digital instruments Nanoscope III) with an etched Si scanning tip.

## RESULTS AND DISCUSSION

Figure **1** shows the XRD pattern for a sample with 100 nm MgO grown on Si(100) substrate with 300 nm thick $SiO_2$ native oxide layer. It can be seen that XRD identify a mixed (200) and (220) orientations of the MgO film. The relative intensity for the reflection from (200) plane is about twice that of (220) plane indicating that the (200) is the predominant orientation. Since the MgO (220) plane has the lowest surface energy of MgO, it should be the preferred orientation. However, in the present case since the substrate is (100) oriented, the interaction between the MgO film and Si substrate leads to dominant (200) orientation of the MgO film. On the other hand the occurrence of (220) peak for the MgO thin films could be attributed to the presence of amorphous $SiO_2$ layers. The occurrence of predominant (100) texture in the MgO film is encouraging because of its importance in achieving high TMR ratios of the device [4,5].

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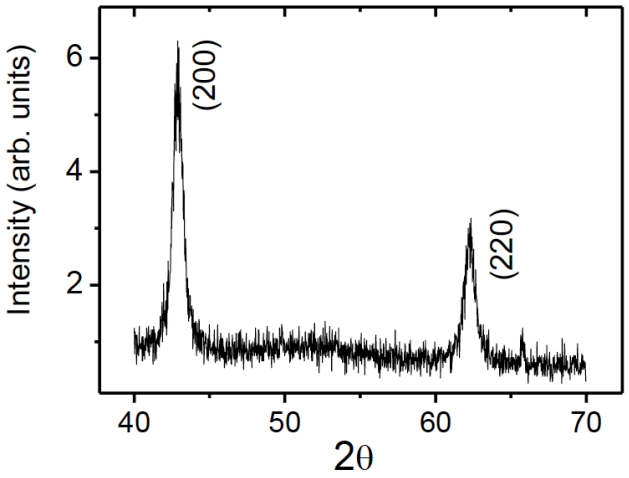


**Figure 1:** XRD patterns of a 100 nm MgO film deposited on Si (100)/$SiO_2$ substrate at room temperature and annealed at 200 °C after deposition.

In first step we vary the substrate temperature while keeping all the other factors constant. Figure **2** shows the two dimensional AFM images of three samples with 1 nm MgO thickness grown at different substrate temperatures: (a) 27 °C, (b) 60 °C and (c) 100 °C. These samples are named as sample-1, sample-2 and sample-3 respectively. Temperatures exceeding 100 °C were not used in the present study in order to avoid cluster formation during the growth of MgO film [9]. No annealing before and after the deposition was carried out for these samples. As shown in Figure **2d**, increase in the growth temperature from 27 °C (room temperature (RT)) to 100 °C causes the rms roughness to increase up to twice its value at RT. This increase in the rms roughness with increasing temperature is attributed to the aggregation of MgO into large islands [8]. This effect is more prominent with increasing temperature due to the removal of adsorbed gases from the surface. For ultra thin films the aggregation of MgO results in nonuniform morphology and increased rms roughness. The observed increase in rms roughness may also originate due to some contamination of MgO surface during sample transport for AFM measurements [10]. In order to get better quality of the films the mobility of the atoms should be reduce during the deposition process. Hence, we propose RT as the suitable growth temperature for the MgO films. However, the values of rms roughness obtained for the three samples in Figure **2a-c** are about 2 to 3 times greater than the atomic spacing of MgO (0.21 nm). Thus the films are far from being atomically smooth and need optimization of other growth parameters. It can be seen from Figure **2a-c** that the

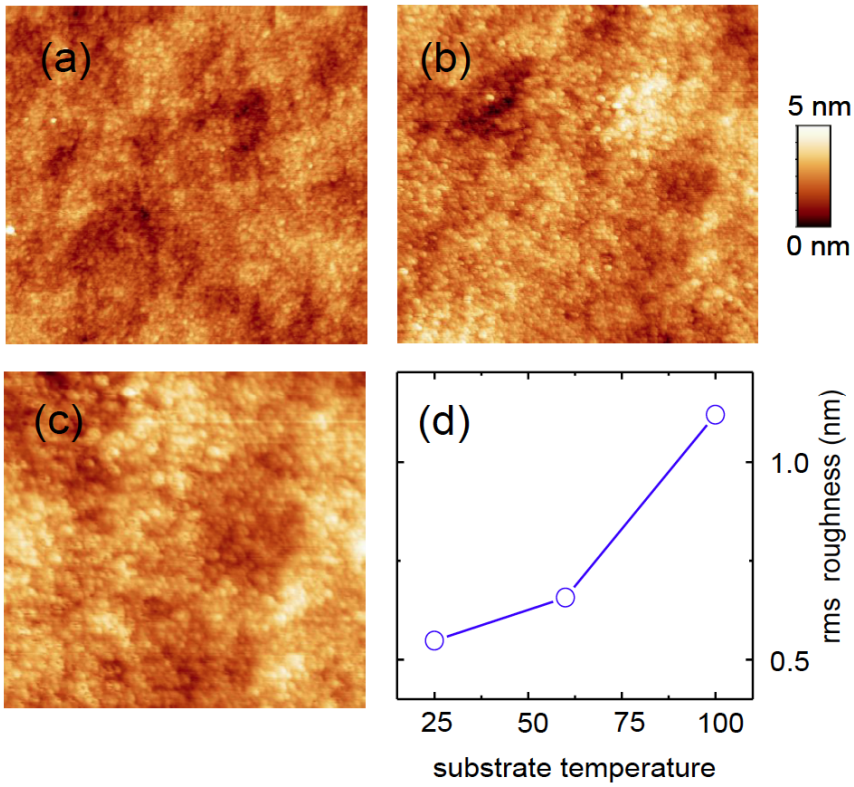


**Figure 2:** The role of substrate temperature for the growth of MgO films. AFM images of 1 nm MgO film grown on $Si/SiO_2$ at (**a**) room temperature and at a substrate temperatures of (**b**) 60 °C, (**c**) 100 °C. (**d**) Dependence of rms roughness on substrate temperature.

average grain size increases slightly with increasing substrate temperature. The effect is not drastic because of the narrow range of substrate temperatures used. However, the increase in grain size with substrate temperature is in agreement with the literature [8, 10, 11]. This effect could be understood by the common nucleation theory [11]. For low temperature growth the mobility of the atoms on the substrate is very low. This results in small nucleation site which leads to small grain size. On the other hand at higher deposition temperature the improved surface mobility of the atoms ensure a large nucleation site, which then generate a bigger sized grain.

In a next step we attempt to improve the rms roughness of the deposited films by annealing the samples after the deposition. The post deposition annealing was performed in-situ for temperatures ranging from 100 C to 400 C for 1 – 2 hours (stabilized at peak temperature). For this purpose the samples were mounted on a heating stage of the molybdenum sample holder equipped with a tungsten filament. At the end of the annealing period the substrates were allowed to cool to RT in the UHV conditions. The surface morphology was then examined *ex-situ* with an AFM. Figure **3** a shows two dimensional AFM images of (a) as-deposited (b) annealed at 100 °C (for 1 hour) and (c) annealed at 200 °C (for 1 hour) samples. The image in Figure **3a** is of sample-1 whereas images in Figure **3b** and **c** are of sample-4 and sample-5, respectively. The three dimensional 2 μm × 2 μm × (~3.5) μm AFM images are also shown in the corresponding insets (Figures **3a-b**). Little change in the surface morphology of the MgO crystal was observed following annealing at 100°C (Figure **3b**) and 200°C (Figure **3c**) for 1 hour. However, a slight decrease (~10 %) in the rms roughness was observed with increasing annealing temperature to 200 °C (Figure **3d**). The decrease in rms roughness with increasing annealing temperature indicates that the surface quality of the films increases on thermal annealing. This effect is attributed to the removal of surface defects upon thermal treatment. Annealing experiments at higher temperatures and longer times result in substantial degradation of the surface quality with some bubble shape structures appearing on the surface. Thus we decided to anneal the samples at 200 °C for 1 hour. This will not only ensure optimum value

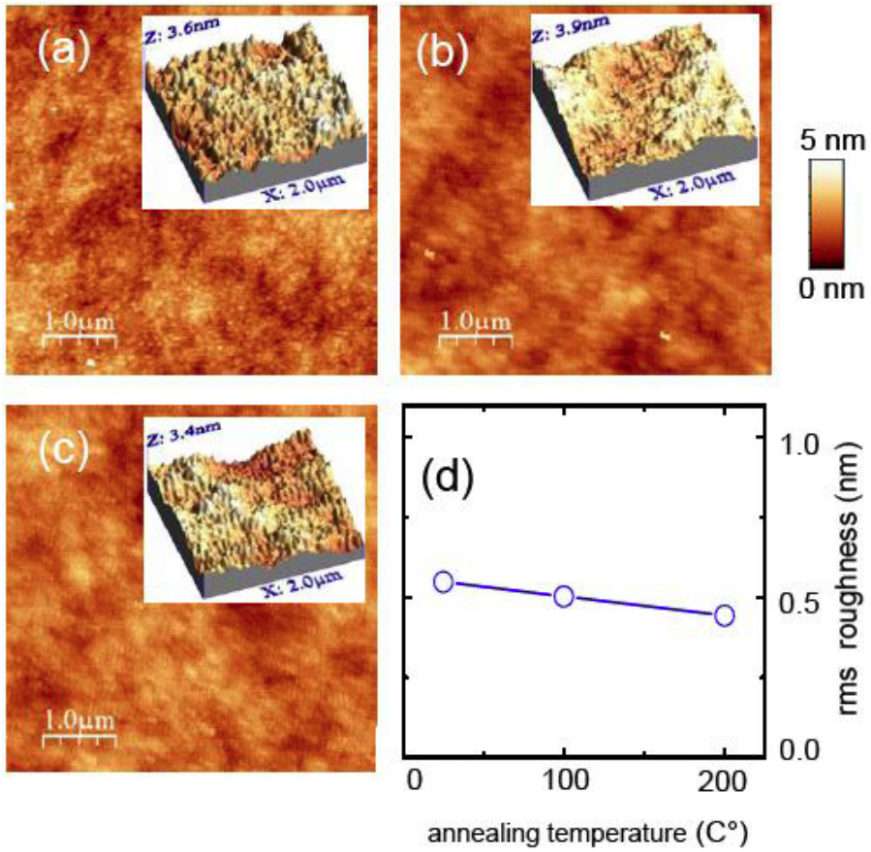


**Figure 3:** The role of post deposition annealing for the growth of MgO films. AFM images of (**a**) as-deposited (**b**) annealed at 100 °C (for 1 hour) and (**c**) annealed at 200 °C (for 1 hour) samples. For each case the film thickness is 1 nm which is grown at room temperature. (**d**) Dependence of rms roughness on annealing temperature.

**Table 1:**

| | Sample-1 | Sample-2 | Sample-3 | Sample-4 | Sample-5 |
|---|---|---|---|---|---|
| MgO Thickness | 1 nm | 1 nm | 1 nm | 1 nm | 1 nm |
| Growth temperature | RT | 60 ˚C | 100 ˚C | RT | RT |
| Post annealing temperature | none | none | none | 100 ˚C 60 min. | 200 ˚C 60 min |
| rms roughness | 0.548 nm | 0.657 nm | 1.33 nm | 0.503 nm | 0.443 nm |

of rms roughness but also eliminate impurities especially water during the annealing process. Additionally, long annealing cycles at high temperatures are also not suitable for devices due to interdiffusion at the interfaces. A summary of all the studied samples is given in Table **1**.

## CONCLUSION

To summarize, we optimized the growth MgO films with minimum surface roughness on $Si/SiO_2$ substrates and on isolated graphene sheets (on $Si/SiO_2$ substrates) using molecular beam epitaxy (MBE). These films were characterized ex-situ using atomic force microscopy (AFM), and X ray diffraction (XRD). Different substrate temperatures and annealing conditions were used for the growth of MgO films. Our results demonstrate that high quality MgO films can be grown at room temperature using molecular beam epitaxy. Post deposition annealing leads to rms roughness better than 0.5 nm without any indication of aggregation or nonuniformaty of film surface. We believe that our MBE frown MgO films are suitable for applications as tunnel barriers in spin-valve devices.

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