Growth and morphology of the epitaxial Fe(110)/MgO(111)/Fe(110) Trilayers

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Abstract

Growth and surface morphology of epitaxial Fe(110)/MgO(111)/Fe(110) trilayers constituting a magnetic tunnel junction were investigated by low-energy electron diffraction (LEED) and scanning tunneling microscopy (STM). STM reveals a grain-like growth mode of MgO on Fe(110) resulting in dense MgO(111) films at room temperature as well as at 250 °C. As observed by STM, initial deposition of MgO leads to a partial oxidation of the Fe(110) surface which is confirmed by Auger electron spectroscopy. The top Fe layer deposited on MgO(111) at room temperature is relatively rough consisting of clusters which can be transformed by annealing to an atomically flat epitaxial Fe(110) film.

Keywords: MgO; Fe(110); LEED; STM

1. Introduction

During the last decade magnetic tunnel junctions (MTJs) consisting of two ferromagnetic electrodes separated by a thin insulating barrier have attracted an increasing interest due to their potential applications in magnetoelectronic devices [1–3]. MTJs consisting of amorphous aluminium oxide (AlO_x) tunnel barrier and ferromagnetic electrodes with high spin polarization [4–7] show relatively low tunneling magnetoresistance (TMR) ratio, up to about 70% at room temperature (RT), which seriously limits the implementation of such structures in spintronic devices. On the other hand, first-principle theories predicted an extremely high MR ratio (over 1000%) for epitaxial Fe(100)/MgO(100)/Fe(100) MTJs with abrupt interfaces between MgO and Fe [8,9]. Since then, experimental evidences for high TMR values at RT have been re-

ported in epitaxial [10] and textured Fe(100)/MgO(100)/Fe(100) MTJs prepared by sputtering [11] as well as in MTJs with amorphous CoFeB electrodes [12]. Very recently, TMR values as high as 410% at RT were demonstrated in epitaxial Co(100)/MgO(100)/Co(100) system [13].

Along with the (100)-oriented Fe films, Fe(110) films can be an interesting candidate as electrode material for MTJs due to the high spin polarization observed on the Fe(110) surface [14]. Recently, TMR values of about 28% at RT have been reported for fully epitaxial Fe(110)/MgO(111)/Fe(110) MTJs prepared by molecular beam epitaxy (MBE) [15,16]. Therefore, the investigation of growth and interface properties is crucial for improving of the performance of MTJs on the basis of Fe(110)/MgO(111)/Fe(110).

In this work we report on growth, morphology, and electronic properties of epitaxial Fe(110)/MgO(111)/Fe(110) trilayers constituting a TMR element which were investigated by means of low-energy electron diffraction (LEED), scanning tunneling microscopy (STM), and

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Auger electron spectroscopy (AES). The growth of ultra-thin MgO layers on the Fe(110) surface at RT as well as at elevated temperature was investigated in detail for different MgO coverages. The effect of annealing treatment on the morphology of the top Fe layer deposited on MgO barrier layer was also investigated.

2. Experimental

All metallic layers as well as the MgO layer were grown by MBE without breaking the ultra-high vacuum (UHV) conditions at any stage of the multilayer sample preparation. Metals (Mo, Fe) were deposited from high purity rods heated by electron bombardment and MgO was evaporated directly from a tungsten crucible also heated by electron bombardment.

All measurements were carried out at RT in a UHV chamber with a base pressure of 8×10^{-11} mbar. STM measurements were performed using an Omicron AFM/STM microscope with polycrystalline tungsten tips which were electrochemically etched *ex situ* and cleaned *in vacuo* by Ar^+ sputtering. The presented STM topographic data were plane fitted to compensate the misalignment of tip and sample. All AES spectra were recorded in the dN/dE mode with 2.5 keV primary electron energy and peak-to-peak modulation voltage of 2 V.

3. Results and discussion

Fig. 1a shows an STM image of the clean atomically flat surface of a 200-Å-thick Fe(110) film deposited at RT on a 100-Å-thick Mo(110) buffer layer on the $\text{Al}_2\text{O}_3(11\bar{2}0)$ substrate and subsequently annealed at about 500 °C to improve the bulk and surface structure. The characteristic height of monoatomic Fe(110) steps was found to be 2.05 Å (see corresponding height profile A) which is in good agreement with the ideal Fe(110) step height of 2.025 Å. A LEED image of the Fe(110) surface (inset in Fig. 1a) shows very sharp spots with a two-fold symmetry typical for bcc Fe(110) surface.

Fig. 1b shows an STM image of the Fe(110) surface after the deposition of nominally 1.2 Å MgO. Roundly shaped MgO clusters with lateral size of 3–8 nm uniformly distributed on the Fe(110) surface can be observed. The height of the MgO clusters varies slightly upon changing the bias voltage achieving saturation about 3 V. A comparable effect has been already observed by Valeri et al. [17] for ultra-thin MgO films on the Ag(001) surface. In the present study the height of MgO clusters was measured at the bias voltage of 3 V to be in the range of 2–6 Å. Fig. 1c represents the surface morphology of a nominal 3-Å-thick MgO film deposited at RT on the Fe(110) surface. The MgO film has now become very dense with the MgO clusters located close to each other, however, without any visible tendency to coalesce into a flat MgO film. The

Fig. 1. $100 \times 100 \text{ nm}^2$ STM images of: (a) 200-Å-thick Fe(110) film surface ($U_T = 1 \text{ V}$; $I_T = 0.12 \text{ nA}$), (b) nominally 1.2 Å ($U_T = 3.1 \text{ V}$; $I_T = 0.12 \text{ nA}$), (c) nominally 3 Å ($U_T = 3.4 \text{ V}$; $I_T = 0.14 \text{ nA}$), and (d) nominally 25 Å ($U_T = 3.5 \text{ V}$; $I_T = 0.13 \text{ nA}$) of MgO on the Fe(110) surface deposited at RT. Profiles A and B correspond to the STM height profiles along the lines shown in (a) and (b), respectively. Insets in (a) and (d) show LEED patterns of the pure Fe(110) surface ($E_{\text{kin}} = 90 \text{ eV}$) as well as of a 30-Å-thick MgO(111) layer ($E_{\text{kin}} = 101 \text{ eV}$), respectively.

density of the MgO clusters and the resolution limitations due to the finite STM-tip curvature do not allow an identification of the Fe(110) surface which could still be present between the MgO clusters. Finally, Fig. 1d shows a surface morphology of a nominal 25-Å-thick MgO film deposited at RT on Fe(110). The dense MgO film consists of round or slightly elliptical clusters. The peak-to-peak roughness was measured to be 6–7 Å on the lateral scale of 100 nm. The fact that stable STM measurement could be performed even at 25-Å-thick films can be possibly attributed to the high defect density in the MgO(111) layer due to the strong stress at the MgO/Fe interface leading to a considerably reduced band gap and thus to a low tunneling barrier height [18]. After the deposition of nominal 30 Å of MgO a well ordered hexagonal (1×1) LEED pattern corresponding to the MgO(111) layer was observed (inset in Fig. 1d).

The growth mode of MgO on Fe(100) has already been extensively studied confirming the layer by layer MgO growth on Fe(100) at RT [19–21]. In contrast, we show that the growth of MgO(111) on Fe(110) follows a Volmer-Weber three-dimensional (3D) growth mode. This result can be expected from the large mismatch between Fe(110) and MgO(111) which is equal to 3.8% along the

(001) direction of Fe and 27.2% along the $\langle 1\bar{1}0 \rangle$ direction of Fe, respectively. On the other hand, the MgO(111) surface is a perfect polar surface with a succession of purely anionic and cationic crystal planes, presenting a theoretical infinite surface energy. A plane MgO(111) surface is thus unstable and is expected to present a rough front growth. Moreover, the recent reflection high energy electron diffraction (RHEED) study confirms the formation of the (111)-oriented MgO 3D islands on Fe(110) [16]. It is interesting to note that MgO(111) occurs in the two mirror related twin variants which preserve the (111) interface plane. Thus, the experiments show that the epitaxial MgO(111) layer is a continuous, though grainy insulating barrier.

In an attempt to produce flat MgO films on the Fe(110) surface, the deposition at 250 °C has been performed. Fig. 2 shows STM images of nominal 10 Å (a) and 15 Å (b) of MgO on the Fe(110) surface deposited at 250 °C which clearly indicates the 3D growth. MgO islands of up to 15 Å can be observed on the Fe(110) surface (Fig. 2a). The peak-to-peak roughness of the 15-Å-thick MgO film was determined to be 10–12 Å on a lateral scale of 100 nm which is considerably higher than observed for the RT growth indicating the advantage of the latter growth procedure. Another possibility for the preparation of the smooth defect-free MgO layer would be the Mg deposition followed by an oxidation step and post annealing. This process in combination with the plasma oxidation was recently shown to deliver better quality tunneling barriers in the MgO-based MTJs [22].

We investigated the MgO growth at RT in more detail and also found that the Fe(110) surface does not remain intact upon MgO deposition. Fig. 3a shows the Fe(110) surface with a nominally 0.3-Å-thick MgO layer on top. Along with the MgO islands small depressions are clearly seen on the Fe(110) surface (dark spots in Fig. 3a). The

Fig. 2. $100 \times 100 \text{ nm}^2$ STM images of the Fe(110) surface after deposition of (a) 10 Å ($U_T = 2.2 \text{ V}$; $I_T = 0.12 \text{ nA}$) and (b) 15 Å ($U_T = 2.7 \text{ V}$; $I_T = 0.12 \text{ nA}$) of MgO at 250 °C. Lower panels represent STM height profiles (A and B) along the lines shown in (a) and (b).

Fig. 3. (a) The Fe(110) surface topography after deposition of nominally 0.3 Å MgO at RT ($U_T = 1.3 \text{ V}$; $I_T = 0.1 \text{ nA}$). Profile A corresponds to the STM height profile along the lines shown in (a). (b) Auger spectra of thin MgO films deposited on Fe(110) surface. The inset in (b) represents a zoom showing the spectra of 50- and 75-Å-thick MgO films.

depth of such depressions was measured to be 0.4–0.6 Å. Most of the depressions were found to be stable upon STM bias voltage variation in the range of 0.5–3.5 V. Such depressions were not observed on the pure Fe(110) surface before MgO evaporation, therefore they are unlikely to be a result of surface contamination and may correspond to partially oxidized Fe(110) surface areas. The question, in which way and to which extent the Fe(110) surface is influenced by the MgO deposition is difficult to answer solely on the basis of STM studies. Fig. 3b represents Auger electron spectra of an MgO/Fe(110) bilayer as a function of MgO film thickness. The intensity of the Fe $M_{2,3}VV$ Auger peak decreases upon MgO deposition, whereas the position of the peak remains almost unchanged. No presence of iron oxide could be detected by means of AES up to 30 Å MgO film thickness. Only at a MgO thickness of more than 50 Å the Fe Auger peak splits into two components. The AES spectra for 50 and 75 Å MgO on top of Fe(110) show peaks (inset in Fig. 3b) which are characteristic of the presence of a significant amount of Fe in an oxidized state [19]. The splitting can be explained by fact that a considerable amount of oxygen had diffused into the Fe layer during growth and formation of Fe–O and Fe–Mg bonds take the place. In this case the observed low-energy peak can be ascribed to formation of thick layer of iron oxide and high energy peak, for example, comes from Fe–Mg bonds, i.e. strong intermixing at interface at such thicknesses of MgO layer is concluded. Generally, the detection of an iron oxide layer at the MgO/Fe interface is very difficult or not possible as the small, shifted component related to the oxidized Fe must be resolved with respect to the large bulk metal contribution. Recently, the oxidation of the Fe(100) surface after MgO evaporation was also concluded from the surface X-ray diffraction [23,24] and vibration spectroscopy [25] data. The observed oxidation of the

Fig. 4. $100 \times 100 \text{ nm}^2$ STM images of the 50-Å-thick Fe top layer: (a) after Fe deposition at RT ($U_T = 4.8 \text{ V}$; $I_T = 0.12 \text{ nA}$) and (b) after subsequential annealing of the system at 300°C ($U_T = 2 \text{ V}$; $I_T = 0.14 \text{ nA}$). Lower panels show STM height profiles (A and B) along the lines shown in (a) and (b). Inset in (b) shows a LEED image of the Fe(110) surface ($E_{\text{kin}} = 78 \text{ eV}$).

Fe(110) surface can lead to imperfections in the tunnel barrier which can influence the transport properties of the MTJs and lower the TMR values [26].

Fig. 4a shows the topography of 50-Å-thick top Fe electrode deposited at RT on the 30-Å-thick MgO barrier layer. The STM image clearly identifies a three-dimensional growth mode of Fe on top of the MgO layer at RT. Roundly-shaped or slightly elongated islands with an average in-plane size of about 10 nm separated by deep trenches are uniformly distributed over the surface. The STM image shows that many of the Fe islands coalesce into bigger islands of undefined shape, however without formation of continuous film. The average peak-to-peak roughness was measured to be 12 Å on the lateral length of 100 nm (see height profile A in Fig. 4a). We found that the crystalline quality of the top Fe layer can be substantially improved by annealing of the trilayer system. Fig. 4b shows an STM image of the Fe surface after annealing the system at about 300°C for 10 min. The Fe surface is now atomically flat with distinct 2.05-Å-high steps confirming the high surface quality of the top electrode. The corresponding LEED image of the top layer Fe(110) surface (inset of Fig. 4b) shows sharp spots with a two-fold symmetry typical for bcc Fe(110). Here it should be mentioned that TMR values of about 28% at RT and 54% at 1.5 K have been reported for fully epitaxial Fe(110)/MgO(111)/Fe(110) MTJs prepared by molecular beam epitaxy (MBE) with both MgO barrier and Fe top electrode deposited at RT without a post annealing step [15,16]. So far annealing of the Fe(110)/MgO(111)/Fe(110) system at about $250\text{--}300^\circ\text{C}$ should improve the crystalline quality of the top Fe layer and possibly its interface with MgO [12] leading as consequence to higher TMR values.

4. Conclusion

We investigated the surface morphology of Fe(110)/MgO(111)/Fe(110) trilayers constituting a MTJ. An island-like growth mode of MgO on the epitaxial Fe(110) layer leading to dense MgO(111) films was revealed by STM. Deposition of MgO at 250°C leads to a substantially higher surface roughness of the barrier layer comparing to the RT growth which may favor pinhole formation. Partial oxidation of the Fe(110) surface upon MgO evaporation was observed by STM and AES which may possibly influence the transport properties of the MTJ. The top Fe layer of high crystalline quality can be fabricated by RT deposition and subsequent annealing of the layered system.

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