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Selective growth of (100)-, (110)-, and (111)-oriented MgO films on Si(100) by pulsed laser deposition

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Selective growth of singly oriented (110)-, (100)-, and (111)-MgO films on Si(100) substrates were obtained by pulsed laser deposition. The effects of deposition temperature, ambient oxygen pressure, and etching of the substrate on the structural properties of the films were studied. It is found that the crystalline orientations of the MgO films are determined at the initial deposition stage by the substrate temperature only. The ambient pressure during deposition and etching of the Si substrates only effect the crystalline quality. Both (110)- and (111)-oriented films show granular grain structures. The (100)-oriented films grown on etched Si substrates display similar granular structures. Those deposited on nonetched Si substrates, however, reveal distinctive columnar grains. The observed phenomena are discussed based on the theory of crystal growth. The mechanism of the orientation selection is attributed to the energy balance between the surface and the interface energies. The varied grain structures are explained by considering the mobility of adatoms in different situations. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1461059]

I. INTRODUCTION

The fabrication of oxide thin-film microstructures on silicon substrates offers tremendous opportunities for developing future microelectronic devices.[1–3] Such microstructures would allow oxide layers to be fully integrated with circuitry. Consequently, the broad electronic, magnetic, and optical properties of oxide materials could be utilized on chip. Unfortunately, direct growth of many these functional oxides on silicon is frequently hampered by extensive interdiffusion or chemical reactions that degrade the properties of the oxides and the underlying silicon. As a solution, the use of thermodynamically stable buffer layers inserted between the functional oxides and the silicon has been proposed. However, the selection and preparation of such a buffer is challenging. The properties of the functional oxides often depend critically on the orientation, surface state, strain, etc., of the buffer layer. Buffer layers with different orientations may even lead to different oxide orientations, and cause large difference in properties due to the high anisotropy of the oxides. Therefore, the controlled orientation growth of buffer layers on silicon is of fundamental importance.

MgO is a highly ionic metal oxide having a NaCl structure and a lattice constant of 4.213 Å. It has been widely used as buffer for growing high-T_C superconducting oxides,[4] ferroelectrics,[5] and colossal magnetoresistive oxides.[6] As a buffer material, MgO has a number of favorable properties. (1) Many perovskite oxides can be epitaxially grown on MgO single crystal substrate. The growth of MgO on silicon therefore provides a template for textured or even epitaxial growth of perovskite oxides. (2) (100)-oriented MgO has a large lattice mismatch of about -9.5% with the (100)-oriented films of most perovskite oxides of typical lattice constant of 3.8 Å. This leads to a large stress in the perovskite oxide films and allows us to study the stress effect in these oxides.[7] (3) As we will show in the present work, three kinds of single-oriented MgO films can be obtained on Si(100). They thus can be used to control the orientations of the perovskite oxides grown on silicon. This is particularly useful if one wants to study anisotropic effects in some of these oxides. (4) Its refractive index of 1.7 is very small compared with other buffer layers such as CeO_2 and YSZ. This makes MgO a suitable buffer layer for growing optical waveguide films. (5) Its low dielectric constant and low dielectric loss make it a good host for microwave devices based on high-T_C superconductors.[8]

Many attempts to grow MgO films on Si(100) by a variety of methods, such as pulsed laser deposition (PLD),[3,7–13] electron-beam evaporation,[14–16] chemical vapor deposition, or metalorganic chemical vapor deposition,[17–19] molecular-beam epitaxy,[20] Sol–gel[21] and atomic layer epitaxy,[22,23] have been reported. Three kinds of single-oriented MgO films, namely (110), (100), and (111) have been demonstrated. In

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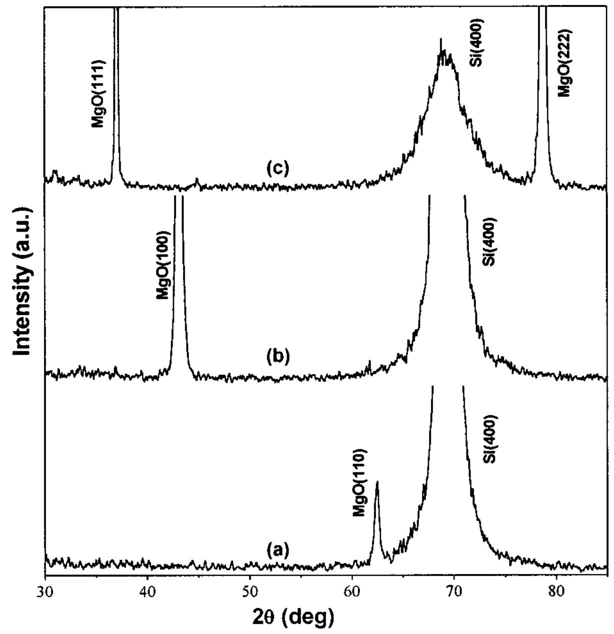

FIG. 1. XRD patterns of MgO films deposited at different initial substrate temperatures on etched Si(100) under an initial ambient oxygen pressure of 1×10^{-5} Torr. (a), (b), and (c) correspond to the initial substrate temperatures of RT, 550 °C and 700 °C, respectively.

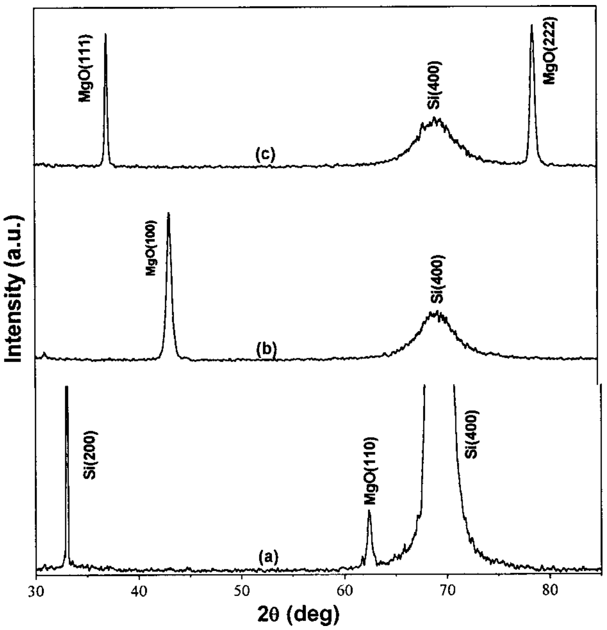

FIG. 2. XRD patterns of MgO films deposited with the same experimental conditions as that in Fig. 1 except that the substrates used here were not etched.

this work, we present the studies on the control of the oriented growth of (110)-, (100)-, and (111)-MgO films on Si(100) by the PLD method.

The basic idea of the orientation control of the film is based on the general theory of film growth. The orientation of the crystalline film is determined, to a large extent, at the nucleation stage of film growth. The deposition temperature plays an important role at that stage. It was found that different deposition temperatures actually lead to different crystalline orientation in the present MgO/Si(100) system. However, if the deposition temperature is too low throughout the film growth process, the resulting films may have very poor crystalline quality or even become amorphous. So a two-step method was used to circumvent the problem. The first step aims to form a seed layer of desired orientation. The second step helps to improve the crystalline quality as the film grows thicker. The effects of the ambient pressure and substrate etching on the crystallinity of the grown films were also studied.

II. EXPERIMENTS

The deposition of MgO films were carried out by PLD (KrF excimer laser, 248 nm wavelength, and 5 Hz repetition rate) in a stainless chamber with base pressure of 2×10^{-6} Torr. The laser fluence was about 2 J/cm^2. The targets were prepared by sintering pellets of ultrafine MgO powder obtained from treated commercial MgO mineral powder with purity of 99.99% by a wet chemical method. The aim of the chemical treatment was to further reduce the size of the powder so that strong and dense MgO targets could be produced. The substrate was positioned parallel to and at a distance of about 4 cm from the target. Two kinds of silicon wafers were used. One was Si(100) covered with native oxides. The other was Si(100) with surface oxides removed by dipping the wafer in 1:20 HF/H$_2$O solution for 5 min.

The total deposition time for each sample was 18 min. During the initial 3 min. which was assigned as the first step, the substrate temperature and the ambient oxygen pressure were set at an initial desired value. The deposition in the remaining 15 min. formed the second step, in which the deposition temperature and the ambient pressure may have been changed to new values. In the present work, three initial deposition temperatures of room temperature (RT), 550 °C and 700 °C and two initial ambient oxygen pressures, 1×10^{-5} and 2×10^{-4} Torr were explored. The substrate temperature in the second step was 650 °C for samples grown at RT initially. Samples grown at the other two initial substrate temperatures were kept at the same initial temperature throughout the second step. The ambient oxygen pressure in the second step was always set at 2×10^{-4} Torr. In order to study the effect of the surface state of the substrate on film growth, etched and nonetched Si(100) substrates were used in parallel for every deposition.

The orientation and crystalline quality of the films were measured by the θ–2θ and ω scans of x-ray diffraction (XRD). The film thickness was measured by an α-step profiler. The surface morphology was examined by atomic force microscopy and scanning electron microscopy (SEM).

III. RESULTS

The deposition rate, obtained by dividing the film thickness by the total number of laser shots used, was about 0.2 Å per pulse. It was almost independent of ambient oxygen pressure and deposition temperature.

Figure 1 shows the XRD patterns of MgO films deposited at different substrate temperatures on etched Si substrate

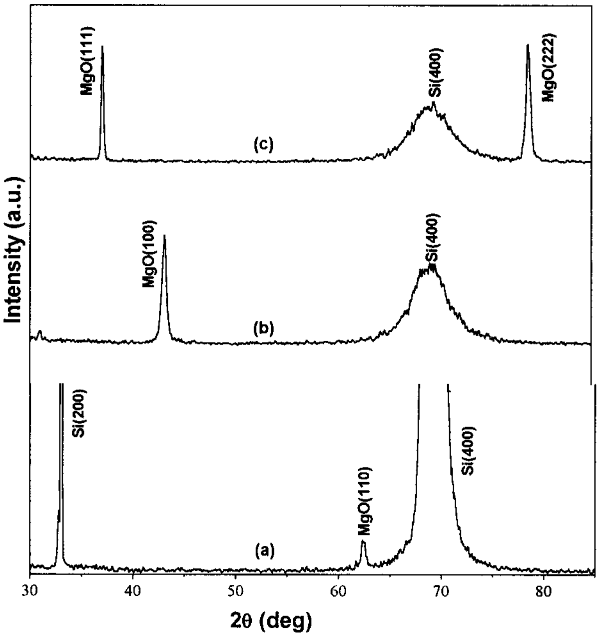

FIG. 3. XRD patterns of MgO films deposited on etched Si(100) under an initial ambient oxygen pressure of 2×10^{-4} Torr. (a), (b), and (c) carry the same meaning as those in Fig. 1.

under an initial ambient oxygen pressure of 10^{-5} Torr. It indicates that the MgO film crystallized in the (110) orientation during the initial deposition at RT, (100) orientation at 500 °C, and (111) orientation at 700 °C.

Figures 2–4 are similar to Fig. 1 but corresponding to the different initial ambient oxygen pressure and substrate surface treatment (etching). Figures 2–4 show that the orientation of the as-grown films was not effected by the ambient oxygen pressure and substrate surface state, but was determined by the deposition temperature during the initial growth stage alone.

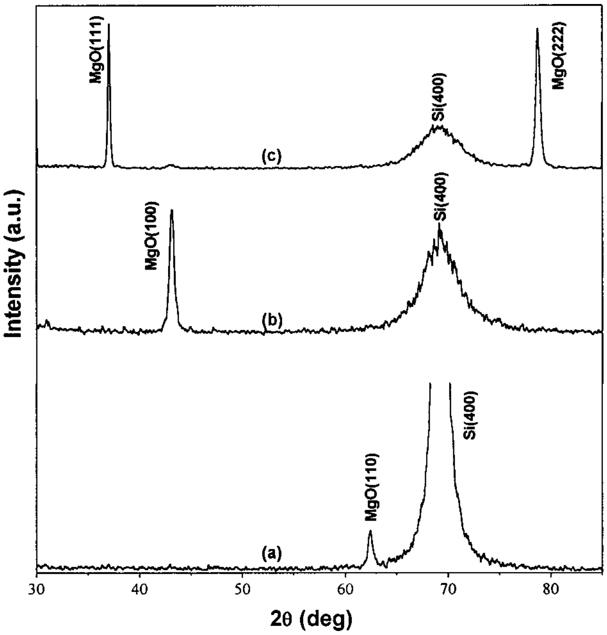

FIG. 4. XRD patterns of MgO films deposited with the same experimental conditions as that in Fig. 3 except that the substrates used here were not etched.

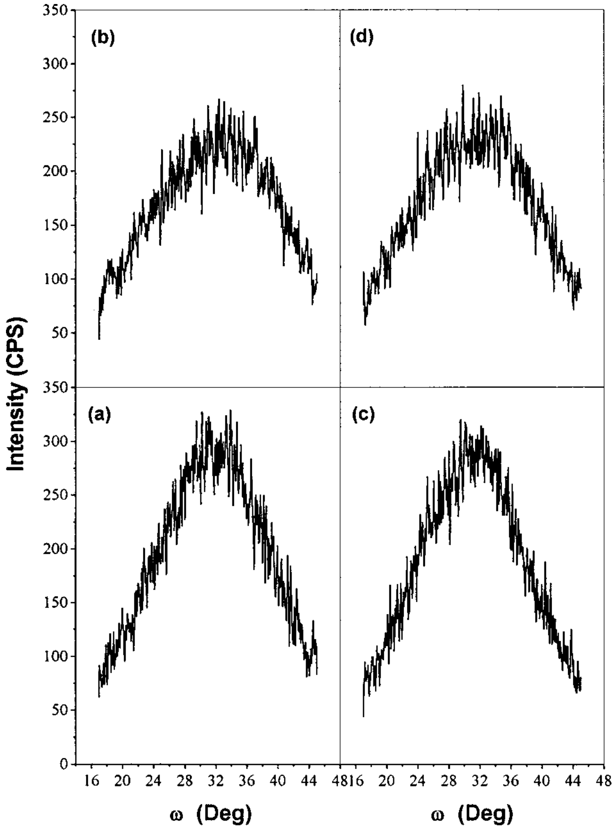

FIG. 5. Rocking curves of the (110)-oriented MgO films prepared under different initial ambient pressure and substrate surface condition. The initial ambient oxygen pressure was 1×10^{-5} Torr for (a) and (c), and 2×10^{-4} Torr for (b) and (d). (a) and (b) correspond to etched substrates and (c) and (d) to the nonetched substrates.

Figures 5–7 are rocking curves of the (110)-, (100)-, and (111)-oriented MgO films prepared at different initial ambient pressures and substrate surface states. For both the (110)- and (100)-oriented films, low initial growth ambient pressure resulted in rocking curves of a narrower full width at half maximum (FWHM). Etching of the Si substrate, on the other hand, had no observable effect on the crystalline quality.

In comparison, the (111)-oriented MgO films show, on the whole, a much narrower FWHM and are less sensitive to the initial deposition pressure than both the (110)-MgO and (100)-MgO films. In addition, etched Si substrates appear to improve the crystalline orientation for (111)-MgO films deposited at the initial pressure of 10^{-5} Torr. These are evident if one compares the FWHM of the rocking curves depicted in Figs. 7(a) and 7(c). For the (111)-MgO films grown at 2×10^{-4} Torr initial pressure, the results are quite the opposite. Etched Si substrates turn out less oriented (111)-MgO films [Fig. 7(a)].

Figure 8 presents the typical AFM images of the three kinds of oriented MgO films. From Fig. 8, it can be seen that the grain size was about 10–20 nm for the (110)-, 100–200 nm for the (100)-, and 300–500 nm for the (111)-oriented films.

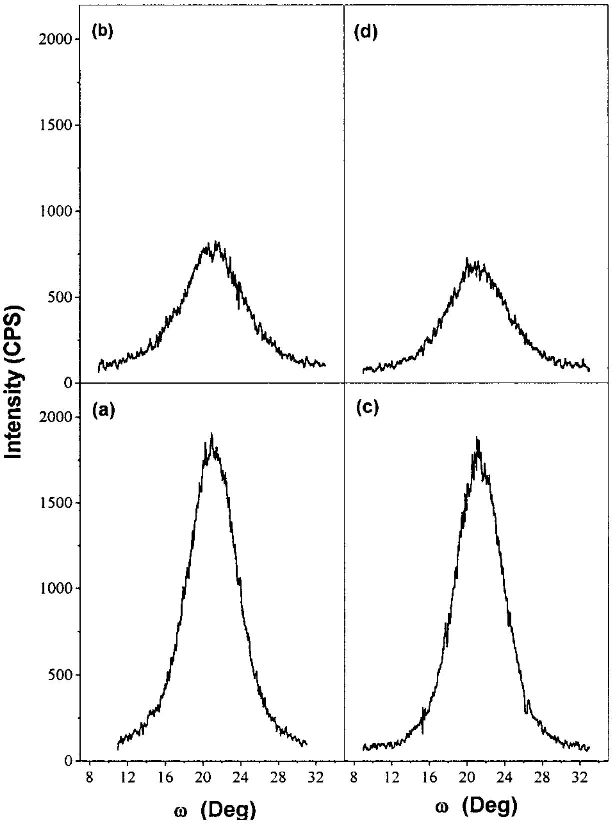

FIG. 6. Rocking curves of the (100)-oriented MgO films prepared under different initial ambient pressure and substrate surface condition. The meaning of the (a)–(d) is the same as those in Fig. 5.

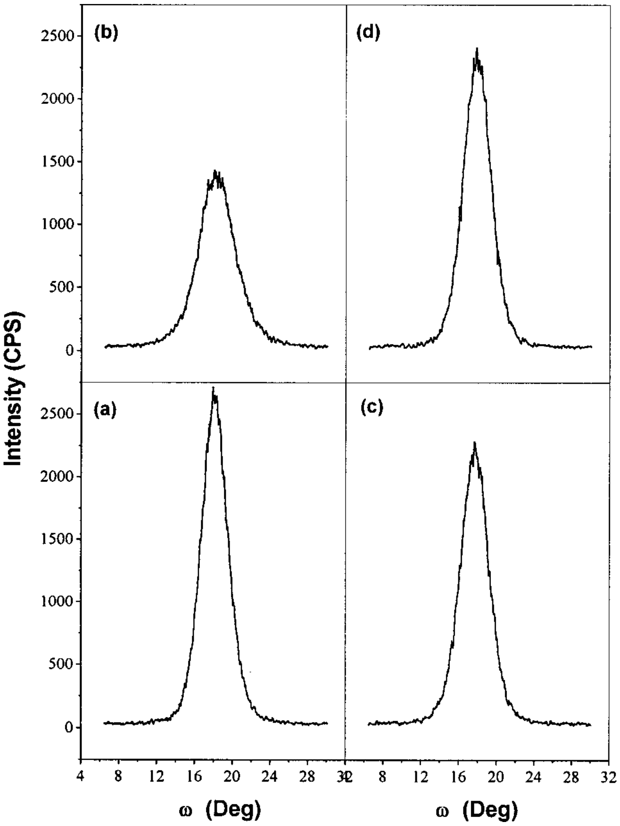

FIG. 7. Rocking curves of the (111)-oriented MgO films prepared under different initial ambient pressure and substrate surface condition. The meaning of the (a)–(d) is the same as that in Fig. 5.

SEM photographs of the cross section of MgO films are shown in Figs. 9–11. The top layers are $LaNiO_3$ thin films. Both the (110)- and (111)-oriented MgO films show granular grain structures, as depicted in Figs. 9 and 10. For the (100)-oriented films, the grain structures appear to be dependent on the surface state of the Si substrates. Those grown on etched Si substrates on the other hand, reveal granular grain structures. However, the film on nonetched Si substrate showed of columnar grain structures. They are shown in Figs. 11(a) and 11(b), respectively.

The surfaces of all three kinds of films were very smooth and devoid of droplets or particulates. As an example, a SEM photograph of the surface of a (100)-oriented MgO film is shown in Fig. 12.

IV. DISCUSSION

A. Nucleation mechanism

1. (110)-oriented films

The growth of (110)-oriented MgO films involves two steps. The first step is deposition at RT for the first 3 min. It is followed by a second step of deposition at 650 °C for 15 min. At the end of the first step, the films are about 18 nm thick. They are amorphous due to the very low substrate temperature. In the time between the first and the second steps, during which the substrates are gradually heated to 650 °C, nucleation, i.e., the formation and growth of crystallites, is homogeneously taking place throughout the volume of the amorphous films. For convenience, the nucleation is denoted as *internal nucleation*. It belongs to the so-called solid–solid transformations, which means that the crystal grows from a solid state.[24] This kind of nucleation has two specific characteristics. One is that the substrate surface has no important effect at all because the nucleation takes place throughout the volume of the amorphous phase. The other is that the mobility of the particles in the solid–solid transformations is limited.[24]

In the second step, during which the substrate temperature is kept at 650 °C, the nucleation and the formation of crystallites can be accomplished soon after the ablated Mg and O species have arrived on the surface of the film. So the nucleation mainly takes place on the film surface. We refer to this process as the *surface nucleation*. It is a kind of heterogeneous nucleation and is dependent on the substrate surface.[24]

The internal nucleation mechanism is more important in determining the orientation and the grain structure of the final (110)-oriented MgO films. The validity of the nucleation mechanism discussed herein is supported by the experimental results on the gain size. The grain size of a film is in general dependent positively on the deposition temperature. For the (110)-oriented films, there are two characteristic temperatures involved. One was RT for the initial 3 min and the other was 650 °C for the remained 15 min. If the grain size was determined during the second step, it should be larger than that of the (100)-oriented films, which were deposited at

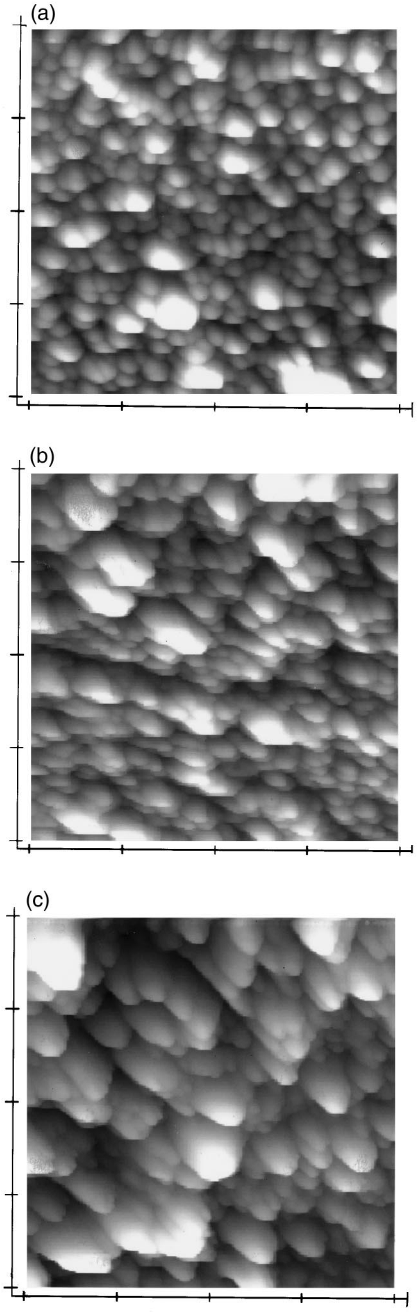

FIG. 8. AFM images of (a) (110)-, (b) (100)-, and (c) (111)-oriented MgO films. Note the actual size is 2×2 μm^2 for (a) and 5×5 μm^2 for (b) and (c).

550 °C for the whole 18 min. However, the grain size (about 10–20 nm) of the final (110)-oriented MgO films is one order of magnitude less than that (100–200 nm) of the (100)-oriented films, as shown in Fig. 8. This indicates that the nucleation and the growth prior to the second step determine the grain structures of the final (110)-oriented films. The remarkably small grain size in (110)-oriented films is a typical characteristic of the solid–solid transformations.

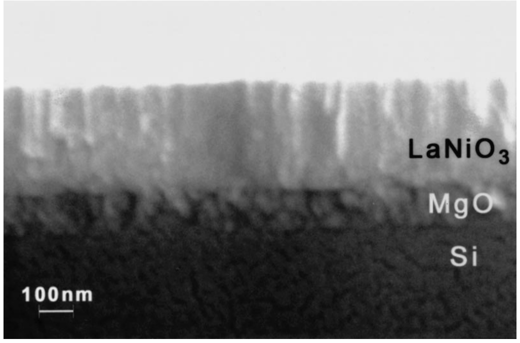

FIG. 9. SEM photographs of cross section of the $LaNiO_3$/(110)-MgO/Si heterostructure. The MgO film was deposited on nonetched Si substrate under the initial ambient pressure of 1×10^{-5} Torr.

2. (100)- and (111)-oriented films

For the (100)- and (111)-oriented films, which were deposited at 550 °C and 700 °C for the whole growing process, respectively, the growth mechanism involves surface nucleation. The resulting grain structures are markedly different from those in (110)-oriented films.

B. Oriented growth

When a material is deposited on a certain substrate surface, a film with preferred orientation is often formed. The orientation is usually temperature dependent. This phenomenon has been observed in a wide range of material systems such as Pt/MgO,[25] YBCO/$SrTiO_3$($LaAlO_3$ or YSZ),[26] Y_2O_3/Si,[27] MgO/Si,[9,10,13–16,18,19] Gd_2O_3/Si,[28] and TiN/Si,[29] etc. However, the relation between the film orientation and the deposition temperature appears to be complex. It was often interpreted by considering the energy balance between interface, surface, and strain energies of the islands during the nucleation stage. Here, we use a similar concept to explain the selective growth of oriented MgO films on Si obtained in our experiments.

From the XRD θ–2θ scan measurements, it is clear that the all three kinds of oriented MgO films have the same lattice constant as that of the bulk MgO. This means that the strain energy is negligible. Therefore, the balance between the surface and interface energy will determine the preferred grain orientation in the present MgO/Si system.

For the (110)-oriented films, the orientation is governed by the internal nucleation mechanism as discussed herein. In this mechanism, only the surface energy is important. How-

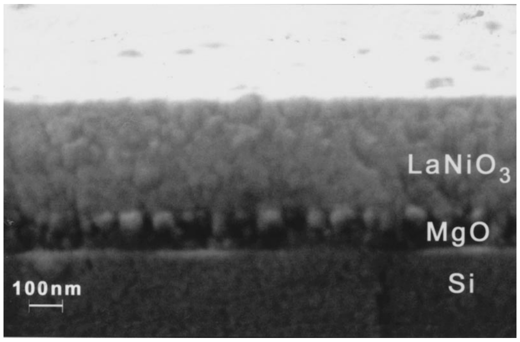

FIG. 10. SEM photographs of cross section of the $LaNiO_3$/(111)-MgO/Si heterostructure. The MgO film was deposited on nonetched Si substrate under the initial ambient pressure of 1×10^{-5} Torr.

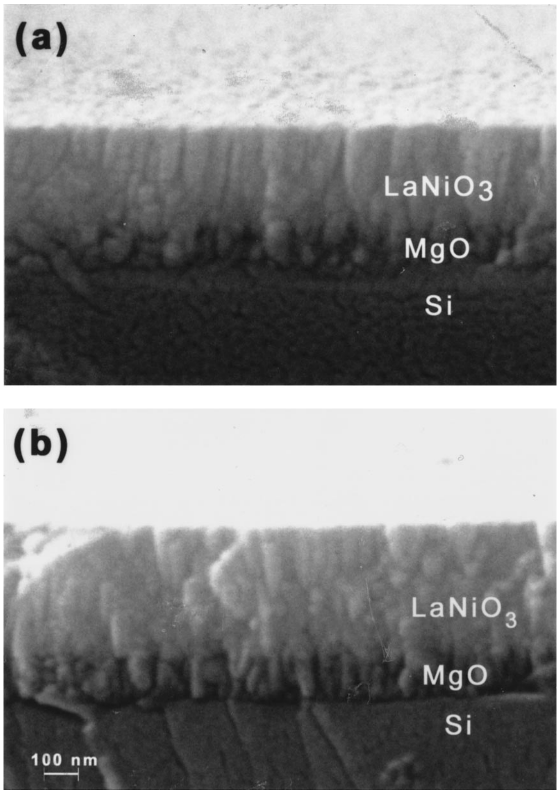

FIG. 11. SEM photographs of cross section of the $LaNiO_3$/(100)-MgO/Si heterostructure. The MgO film was deposited on etched (a) and nonetched Si substrate (b) under the initial ambient pressure of 1×10^{-5} Torr.

ever, for the (100)- and (111)-oriented films, whose orientation is determined by the surface nucleation mechanism, both the interface and the surface energy need to be considered.

The surface energy can be considered as the increase in the potential energies of the surface atoms due to the breaking of bonds to form two separate surfaces. The surface energy is therefore proportional to bond breaking in separating the surfaces.[30] The two-dimensional density of Mg atoms in (100), (110), and (111) surfaces of the MgO crystal is $2/a^2$, $1.41/a^2$, and $2.31/a^2$, respectively, where a is the lattice constant of the MgO crystal. So, the (110) surface involves the least number of broken bonds and thus represents the lowest surface energy. The (111) surface has the largest surface energy and the (100) surface has the intermediate surface energy. Therefore, MgO films grown at RT initially are completely (110) oriented.

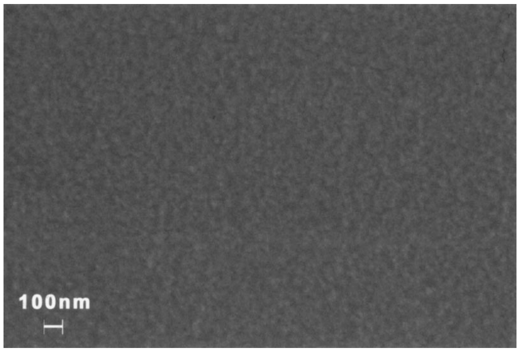

FIG. 12. SEM photographs of a (100)-oriented MgO films deposited on etched Si(100) and under the initial ambient pressure of 1×10^{-5} Torr.

At a high substrate temperature, the Si surface becomes active and reacts easily with oxygen.[31,32] The strong chemical reaction between the Si substrate surface and oxygen atoms absorbed on the substrate surface represents a large interface energy. When the substrate temperature is so high that the interface energy is larger than the surface energy, the interface energy will govern the growth orientation. In the case of MgO/Si growth at substrate temperature of 700 °C, the interface energy is assumed to be larger than the surface energy. Consequently, the formation of (111)-oriented MgO films is favored. From the atomistic viewpoint, the high temperature will rapidly lead to a substrate surface covered with oxygen even under an ambient oxygen pressure of 10^{-5} Torr. As a result, the high surface mobility of the Mg species allows rapid establishment of stable Mg—O bonds and leads to formation of a densely populated Mg atom plane. It is then followed by a layer of oxygen. Consequently, alternate layers of Mg and O will be grown in succession. This means, once again, a MgO film with the (111) orientation.[22]

At an intermediate substrate temperature of 550 °C, both the surface energy and the interface energy are effective in controlling the crystal growth orientation. The competition between the surface energy and the interface energy will therefore lead to the (100) orientation growth.

C. Crystal quality

The FWHM of a rocking curve describes the spread of the crystallite orientation distribution. It is often used as an index for quantifying the crystalline quality of the films. Figures 5 and 6 show that, for the (110)- and (100)-oriented films, (1) the substrate surface state had no major effect on the crystalline quality of the films, and (2) the high vacuum at the initial growth stage was favorable for improved crystalline quality of the film. For the (110)-oriented films, whose formation is dominated by the internal nucleation mechanism, it is naturally uneffected by the substrate surface state. For the (100)-oriented films, which were deposited at 550 °C, the interaction between the deposited species and the substrate surface is not strong enough to produce an observable effect on the crystalline quality. The dependence of the crystalline quality on the ambient pressure can be attributed to the fact that a higher vacuum helps to retain the kinetic energy of the ablated species during their transportation from target to substrate. These species of high kinetic energy diffuse better on the substrate surface and yield improved crystallites.

For MgO films deposited at 700 °C, however, the etched and nonetched Si substrates produce different crystalline quality. This suggests that at high deposition temperatures the surface state of the substrate does play a significant role in the crystalline film growth. This is because the interaction between the film and the substrate becomes strong due to the high reactivity between deposited the Mg atoms and the Si substrate surface at 700 °C.

Another point for the (111)-oriented MgO films is that their crystalline quality is independent on the growth ambient pressure if Si substrates with native oxides are used. This can be interpreted by considering the interface energy. Si substrates covered by a layer of native oxide produce a very strong interface interaction. Comparing to this strong interaction, the effect of the change of the kinetic energy of the adatoms brought about by varying the ambient pressure is negligible. So the crystalline quality shows no dependence on the ambient pressure.

D. Grain structures

There are two types of grain structures often observed in films. One is the columnar structure and the other is the granular structure. It is generally believed that the formation of columnar grain structures is primarily due to the limited mobility of deposited atoms on the substrate surface. The high mobility of adatoms, on the other hand, leads to a granular grain structure.[33,34] It is noted that this rule can only be applied to films whose formation is governed by surface nucleation. The mobility can be characterized by diffusion constant $D \propto \exp\{-V_s/k_B T\}$, where T is the substrate temperature, V_s is the potential-energy barrier for an adatom to diffuse from site to site and k_B is the Boltzmann constant.[35]

For the (110)-oriented films whose formation is governed by the internal nucleation mechanism, the grain structure is always granular and not dependent on the surface state of the Si substrate. At the deposition temperature of 550 °C, Si substrates with native oxides have stronger interaction with deposited the Mg atoms than the etched Si substrates. This means that V_s is larger on the nonetched Si substrate than on the etched ones. As a result, the columnar structures are formed preferentially on etched Si substrate.

However, the (111)-oriented MgO films grown on either etched or nonetched Si substrates show granular grain structures. This can be attributed to the fact that mobility of Mg atoms on both kinds of Si substrate surface at the high temperature of 700 °C is very large.

V. CONCLUSION

Oriented MgO films were prepared on Si(100) substrate by PLD method. It was found that the orientation selection was controlled solely at the initial stage of deposition by the substrate temperature. Singly (100)-, (100)-, and (111)-oriented films were obtained with initial substrate temperature of RT, 550 °C and 700 °C, respectively. Etching of the Si substrate and the ambient oxygen pressure showed no observable effect on the crystalline film orientation. The mechanism of the orientation selection is attributed to the balance between the surface and the interface energies. The crystalline quality and the grain structures of the three singly oriented MgO films have also been studied. Their features and dependence are explained by considering the nucleation mechanism and the mobility of adatoms under different situations.

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