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Epitaxial growth and characterization of high-quality aluminum films on sapphire substrates by molecular beam epitaxy

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Abstract: High-quality Al epitaxial films with homogeneous thickness have been epitaxially grown on 2-inch sapphire substrates by molecular beam epitaxy with an in-plane alignment of Al[1-10]//Al_2O_3[1-100]. The as-grown about 200-nm-thick Al (111) films grown at 750 ºC show excellent uniform thickness distribution over the whole 2-inch substrates and very flat Al surface with the surface root-mean-square roughness of 0.6 nm, as well as high crystalline qualities with the Al(111) full width at half maximum as small as 0.05°. There is no interfacial layer existing between as-grown Al epitaxial films and sapphire substrates. Instead, sharp and abrupt Al/Al_2O_3 hetero-interfaces are achieved. The effects of the growth temperature on the surface morphologies and the crystalline qualities of as-grown Al epitaxial films have been studied in detail. This achievement of Al epitaxial films is of great importance in the application of Al-based microelectronic devices.

Keywords: Al epitaxial films; molecular beam epitaxy; roughness; full width at half maximum; interfacial layer.

1. Introduction

The growth of Al epitaxial film has been attracted considerable attention due to its superior properties for the application in metal-oxide-semiconductor microelectronic devices, emerging magnetic data storage and subwavelength surface plasmonic devices.[1-9] For these applications, the performance shows clear reliance on the Al film crystalline quality, morphology, and structure of the metal–oxide interfaces.[10-14] On the one hand, a single crystalline Al epitaxial films would be very important, with the full benefit of their outstanding electrical and structural properties compared with those of polycrystalline and amorphous Al epitaxial films.[1-9] On the other hand, the metal/oxide interfaces should also be defect-free and atomically smooth, for the migration of electrons. The recent studies find that these properties are usually limited by the quality of Al epitaxial films,[13-14] thus, the growth of high-quality Al epitaxial films is of great importance. Meanwhile, Al epitaxial films are usually prepared in small-size, which, to some extent, increases the cost of the fabrication of microelectronic devices.[1-14] Therefore, the improvement in quality and size of Al epitaxial films is needed urgently. In this regard, much effort has been made to grow high-quality and large-size Al epitaxial films on 2-inch Si substrates by molecular beam epitaxy (MBE).[17-20] But the quality of as-grown Al epitaxial films is still could not meet the requirement due to the large lattice mismatch between Al epitaxial films and Si substrates.[21-22] On the contrary, sapphire is more appropriate for the epitaxial growth of Al epitaxial films. The lattice mismatch between Al epitaxial films and sapphire substrates is as small as 0.08%, which is beneficial to the nucleation of Al atoms on sapphire substrates and thereby the growth of high-quality Al epitaxial films. In addition, sapphire has excellent physical and chemical stabilities at high temperature,[23-24] which is good for the high-temperature growth of Al epitaxial films for achieving high-quality Al epitaxial films.

In this work, we report on the growth of 2-inch high-quality Al epitaxial films on sapphire substrates by MBE. The surface morphologies and the crystalline qualities for as-grown Al epitaxial films are studied by white-light interferometry, *in-situ* reflection high energy electron diffraction (RHEED), field emission scanning electron microscopy (FESEM), atomic force microscopy (AFM), high-resolution X-ray diffraction (HRXRD), and high-resolution transmission electron microscopy (HRTEM). This work of growing 2-inch high-quality Al epitaxial films is of great importance in the future application of Al-based microelectronic devices.

2. Experimental

The sapphire substrates were taken degassing treatment for 30 min in an ultra-high vacuum (UHV) load-lock chamber with a background pressure of 1.0×10^{-8} Torr. Subsequently, they were transferred into the UHV MBE growth chamber with a background pressure of 2.0×10^{-10} Torr. The sapphire substrates were annealed at 850 ºC for 60 min to remove the surface contaminations. During the growth, high purity N_2 (7N) was supplied through an inert gas purifier with a flow rate of 1 sccm. Solid-state high-purity Al was used as the precursor of Al and the Al comcell was set at 1100 ºC. The rotation rate of 5 round-per-min for the substrates was set as constant to guarantee the homogeneous thickness of Al epitaxial films. The growth temperatures were changed from 550 to 850 ºC to study the surface morphologies and structural properties of as-grown Al epitaxial films. *In-situ* RHEED was used to monitor the growth condition during the whole procedure. The as-grown Al epitaxial films were characterized by white-light interferometry (Y-Wafer GS4-GaN-R-405), *in-situ* RHEED, FESEM (Nova Nano SEM 430 Holland), AFM (MFP-3D-S Asylum, American), HRXRD (Bruker D8 X-ray diffractometer with Cu Kα1 X-ray source λ=1.5406 Å), and HRTEM (JEOL 3000F, field emission gun TEM working at a voltage of 300 kVwith a point to point resolution of 0.17 nm).

3. Results and discussion

The thickness distribution of Al epitaxial films grown by MBE is measured by white-light interferometry. Fig. 1a is a typical photograph of the Al epitaxial films grown on sapphire substrates at 750 ºC, and its corresponding thickness distribution measured by white-light interferometry is shown in Fig. 1b. One can clearly see that 201 nm Al epitaxial films are covering the whole 2-inch substrate uniformly, which reveals very homogeneous Al epitaxial films.

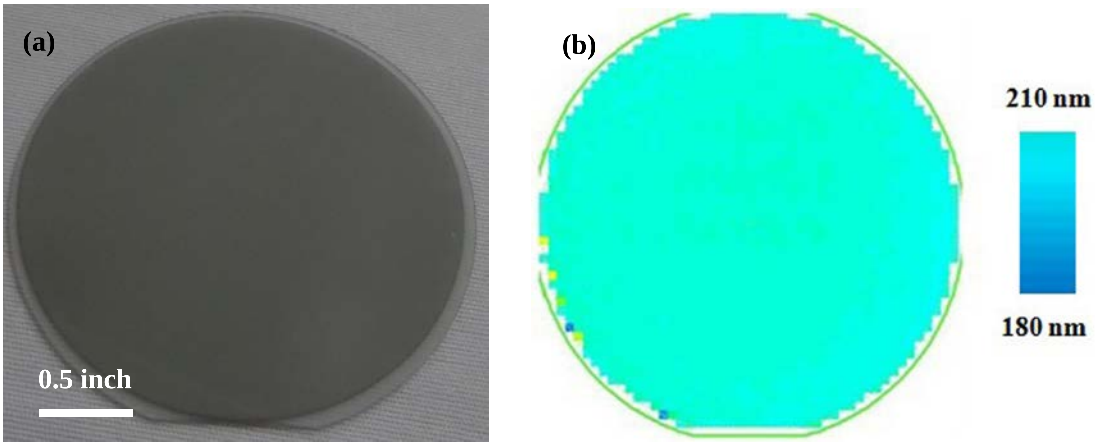


Fig. 1. (a) Typical photograph of the Al epitaxial films grown on sapphire substrates at 750 ºC and (b) its corresponding distribution of thickness.

The *in-situ* RHEED is used to monitor the whole growth process. Fig. 2a illustrates the RHEED patterns for sapphire substrates after 60-minute annealing at 850 ºC. As observed, the RHEED patterns are sharp as well as streaky. This result reveals that residual surface contaminants on sapphire surface have been removed, and very smooth surface has been obtained for the subsequent growth.[25-27] After the following epitaxial growth of ~200-nm-thick Al epitaxial films at 750 ºC by MBE, sharp and clear RHEED patterns for Al epitaxial films can be observed as well, which indicates that single-crystalline Al epitaxial films with a very smooth surface are achieved.[28-29] as shown in Fig. 1b. Furthermore, the RHEED patterned got in this work is much sharper than that grown on Si substrates.[17] In this regard, we can obtain an in-plane alignment of Al [1-10]//Al_2O_3 [1-100] between Al epitaxial films and sapphire substrates by studying the RHEED measurements carefully.[30-31]

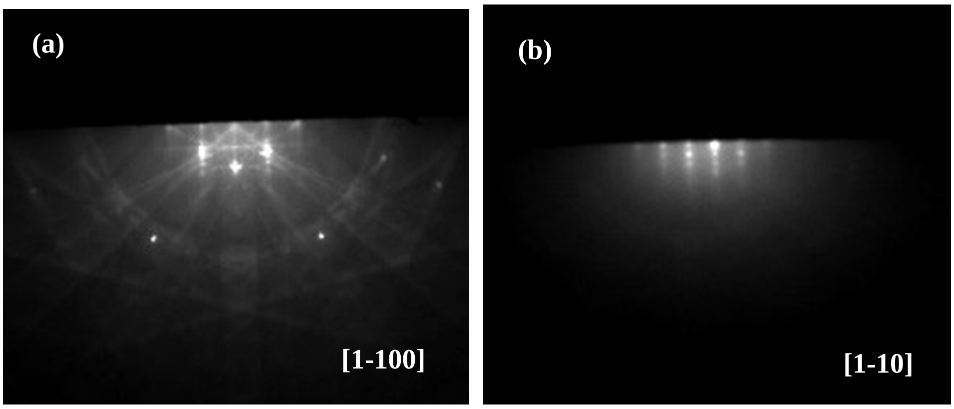


Fig. 2. RHEED patterns for (a) sapphire substrates after 60-minute annealing at 850 ºC, and (b) about 200 nm Al epitaxial films grown at 750 ºC.

The surface morphologies of Al epitaxial films grown at the temperature ranging from 550 to 850 ºC are studied by both FESEM and AFM. Fig. 3a is a FESEM image for ~200-nm-thick Al epitaxial films grown at 550 ºC. There are many island particles distributing on the surface. With the growth temperature increasing, the coalescence of island particles is promoted. Furthermore, if the ~200-nm-thick Al epitaxial films are grown at 750 ºC, one can see very smooth Al surface, as shown in Fig. 3b. However, if the growth temperature further increases to 850 ºC, the Al film surface becomes slightly rough. Meanwhile, the Al epitaxial films thickness is measured to be 201 nm, which agrees well with the value obtained from white-light interferometry measurement, as shown in Fig. 3c.

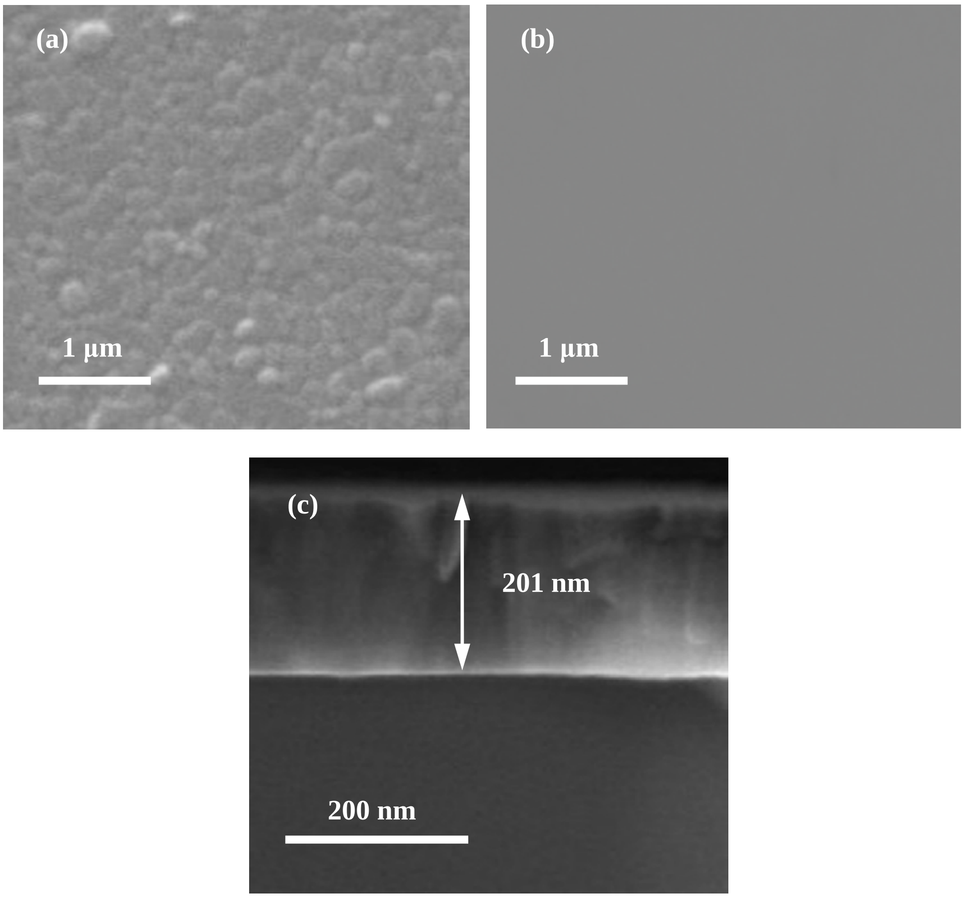


Fig. 3. FESEM images for ~200-nm-thick Al epitaxial films grown at (a) 550 and (b) 750 ºC. (c) Cross-sectional SEM image for ~200-nm-thick Al epitaxial films grown at 750 ºC.

The temperature dependence of surface roughness for ~200-nm-thick Al epitaxial films is characterized by AFM, as shown in Figs. 4a and b. One can clearly find that the root-mean-square (RMS) roughness for ~200-nm-thick Al epitaxial films grown at 550 and 650 ºC are 3.5 and 2.0 nm, respectively. As the growth temperature increases to 750 ºC, the as-grown Al epitaxial films show very smooth surface with a RMS roughness of 0.6 nm, as show in Fig. 4b. These results can be attributed to the difference in diffusion length of Al atoms.[32-33] At higher temperature, the diffusion length of an atom is large, which leads to the formation of smoother surface.[32-35] However, in our case, when Al epitaxial films are grown 850 ºC, the Al surface becomes rough with a RMS roughness of 1.0 nm. This can be explained to the increase of desorption rate of Al films as the increase in growth temperature, which results in slight increase of Al surface roughness.[36-38] Furthermore, the RMS roughness for commercially available Al (111) grown by Czochralski method is in the range of 3-5 nm. These results indicate the falter Al surface grown on sapphire substrate by MBE.

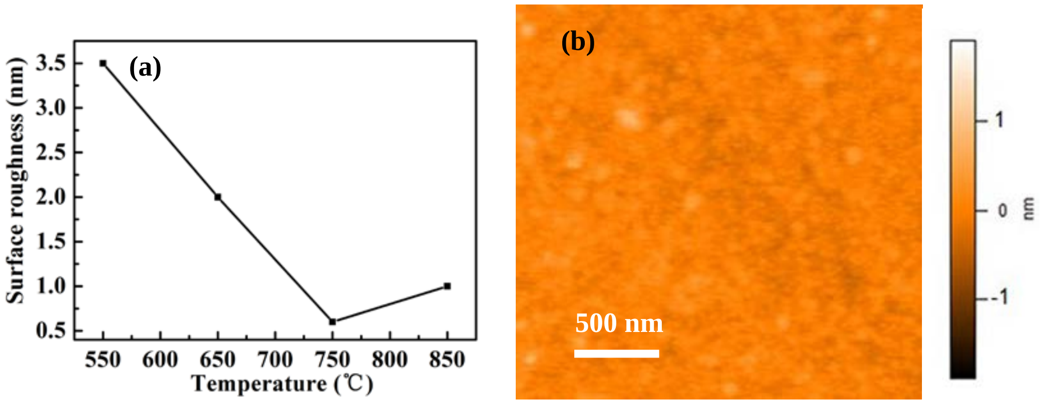


Fig. 4. (a) AFM images for 200-nm-thick Al epitaxial films grown at 750 ºC. (b) Temperature dependence of RMS roughness for Al epitaxial films grown on sapphire substrates at temperatures ranging from 550 to 850 ºC.

The structural properties of Al epitaxial films grown on sapphire substrates are studied by HRXRD. Fig. 5a is a typical 2θ-ω scan for Al (111) films grown on sapphire substrates. The peaks observed at 2θ=38.6° and 2θ=82.5° are attributed to Al (111) and Al (222), respectively;[39] while the peaks found at 2θ=41.7° and 2θ=90.5° are ascribed to Al_2O_3 (0006) and Al_2O_3 (00012), respectively.[40] Therefore, single-crystalline Al epitaxial films have been grown on Al_2O_3 substrates with the out-of-plane epitaxial relationship of Al (111)//Al_2O_3 (0001). Fig. 5b shows the φ scan for Al_2O_3 (1-102), where we can see the three-fold rotational peaks with an interval of 120°. It also reveals φ scan for Al (1-13), from which one can clearly identify six-fold rotational peaks with an interval of 60°. In this regard, we obtain the in-plane epitaxial relationship of Al [1-10]//Al_2O_3 [1-100] with a lattice mismatch of 0.08%, which is consistent well with the RHEED measurement.[41-43]

The X-ray rocking curve is deployed to further investigate the crystalline quality of as-grown 200-nm-thick Al epitaxial films. Fig. 6a is the X-ray rocking curve for ~200-nm-thick Al epitaxial films grown on sapphire substrate at 750 ºC, which shows the full-width at half-maximum

(FWHM) of Al (111) is as small as 0.05°. The FWHM for commercially available Al (111) substrates with the thickness of 1.0 mm grown by Czochralski method is as large as 0.5° got from the Hefei Kejing Materials technology Co., Ltd, American MTI corporation.[39] Meanwhile, the FWHM for Al films grown on Si substrates also lager than that obtained in this work.[17-18] The possible reason might lie in the larger lattice and thermal expansion coefficient mismatches between Al and Si,[17-20] which would lead to the formation of high stress and high dislocation density in the Al films and thereby result in poorer quality of Al films.[17-22] In order to circumvent the issue, several technologies, such as buffer layer, epitaxial lateral overgrowth, pattern design of the Si substrates, *etc*, need to be used.[44-51] These technologies can reduce dislocation density and release the stress to some extent, and eventually benefit the growth of high-quality Al films on Si substrates.[44-51] Therefore, these evidences show the higher crystalline quality of Al epitaxial grown in this work. This achievement of higher quality of Al epitaxial films mainly may be ascribed to two aspects. One is the very small lattice mismatch between Al epitaxial films and sapphire substrates, the other is the suitable growth temperature. The former is beneficial to the nucleation of Al epitaxial films and leads to the formation of few dislocations, and thereby enhances the growth of higher-quality Al epitaxial films. The latter provides enough energy for the migration of Al precursors on the substrate surface, enhances the growth process, and eventually leads to the growth of higher-quality Al epitaxial films.

The temperature dependence of FWHM for ~200-nm-thick Al (111) is also studied, as show in Fig. 6b. One can clearly find that as the growth temperature increases from 550 to 750 ºC, the FWHM for Al epitaxial films narrows gradually. This means that the crystalline quality of as-grown Al epitaxial films improves accordingly. We attribute this to the increase in diffusion length of Al atoms, which may enhance the annihilation and coalescence of dislocations, and thereby improve the crystalline quality of Al epitaxial films eventually.[32-35] However, if the growth temperature further increases, the crystalline quality of Al epitaxial films becomes poorer with the broadened FWHM. This can be explained to the higher growth temperature, which leads to the larger Al desorption rate, and may result in slightly poorer crystalline quality of Al epitaxial films ultimately.[36-38]

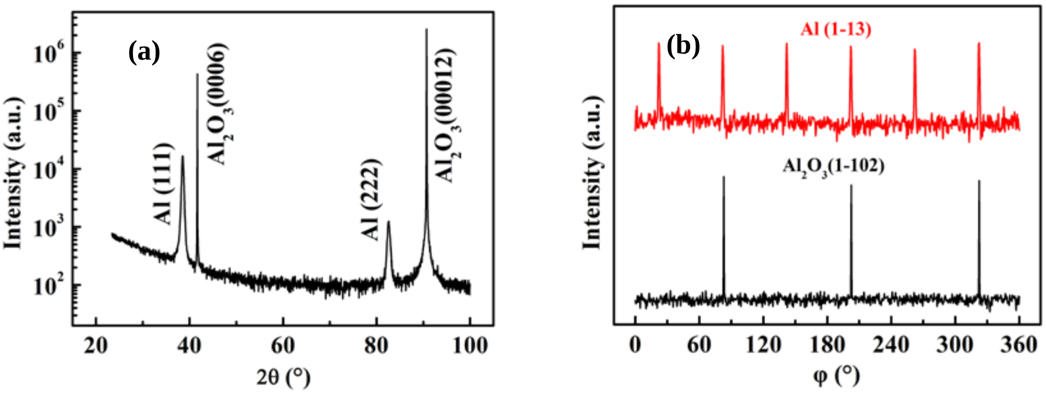


Fig. 5. Typical (a) 2θ-ω scan for Al (111) films grown on sapphire substrates, and (b) φ scans for Al (1-13) and Al_2O_3 (1-102).

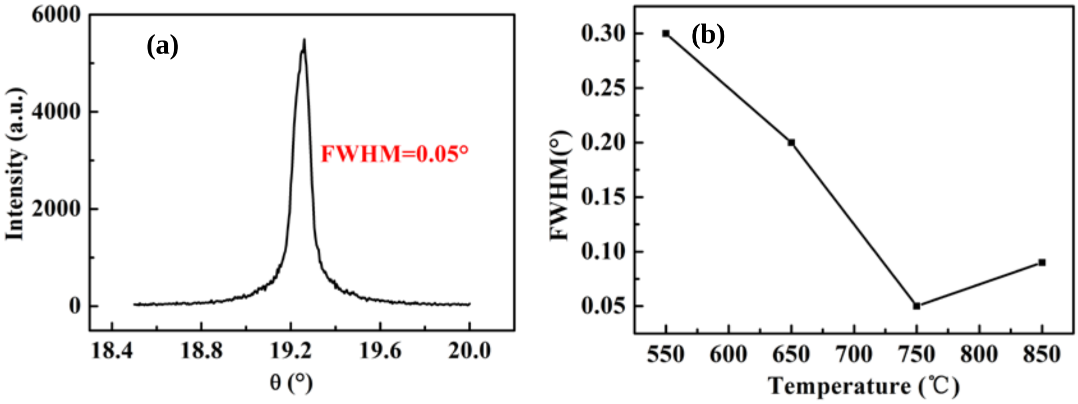
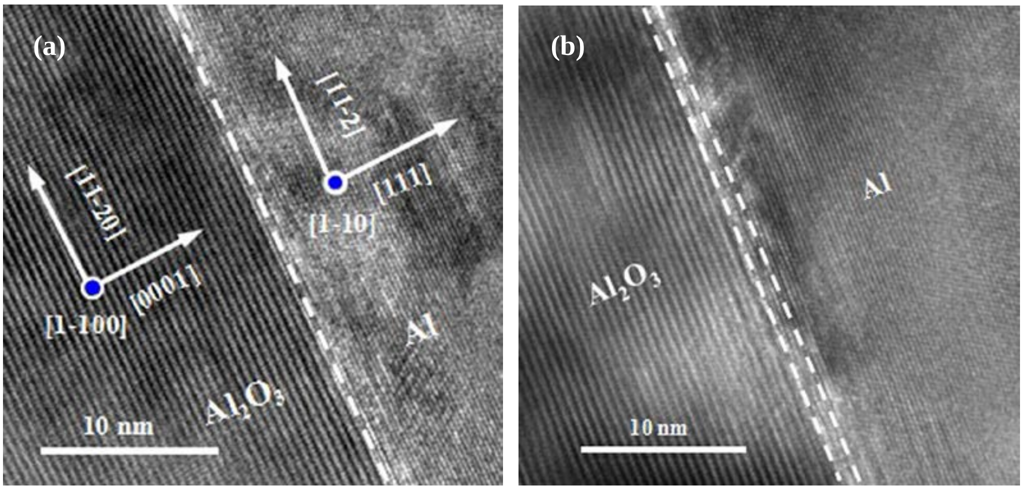


Fig. 6. (a) X-ray rocking curve for 200-nm-thick Al (111) grown at 750 ºC. (b) Temperature dependence of the FWHM for Al epitaxial films grown on sapphire substrates at temperatures ranging from 550 to 850 ºC.

The interfacial properties of Al/Al_2O_3 hetero-interfaces are characterized by cross-sectional TEM and selected area electron diffraction (SAED), as shown in Figs. 7a-d. Fig. 7a is a cross-sectional TEM image with high-magnification for Al/Al_2O_3 hetero-interfaces grown at 750 ºC, from which one can clearly identify that there is no interfacial layer existing between Al epitaxial films and sapphire substrates. Instead, sharp and abrupt interfaces are observed,[52-54] which presents a striking contrast to that grown by other groups with a thick interfacial layer.[1, 55] Fig. 7c is the SAED image for Al/Al_2O_3 hetero-interfaces grown at 750 ºC, where we can obtain the epitaxial alignments between Al and Al_2O_3 are of Al (111)//Al_2O_3 (0006) and Al [11-2]//Al_2O_3 [11-20]. Furthermore, the in-plane alignment of Al [1-10]//Al_2O_3 [1-100] can be deduced from the direction of the electron diffraction and is confirmed by the φ scans.[56-58] In this case, these results once again confirm the high-quality Al epitaxial films have been grown on sapphire substrates. However, Al films grown at higher temperature of 850 ºC show an interfacial layer thickness of 1.0 nm, as shown in Fig. 7b.

The temperature dependence of interfacial layer thickness between Al epitaxial films and sapphire substrates is shown with temperatures ranging from 550 to 850 ºC, Fig. 7d. We can find that the interfacial layer thickness is 2.5 nm at the growth temperature of 550 ºC, and gradually decreases to 0 nm as the growth temperature increases to 750 ºC. However, when the growth temperature further increases, the interfacial layer thickness increases to 1.0 nm. The formation of the interfacial layer may ascribed to interfacial reactions between the Al epitaxial films and sapphire substrates. As we know that there are many dislocations exist in the interfacial layer, which may propagate into the subsequent films and deteriorates the quality of as-grown films. In this case, the tendency of temperature dependence of interfacial layer thickness agrees well with that of crystalline quality. Furthermore, the sharp and abrupt interface is of great importance for the application of Al-based devices. Future work should be focused on the preparation of high-quality oxide films and semiconductor films in turn on this high-quality Al epitaxial films.



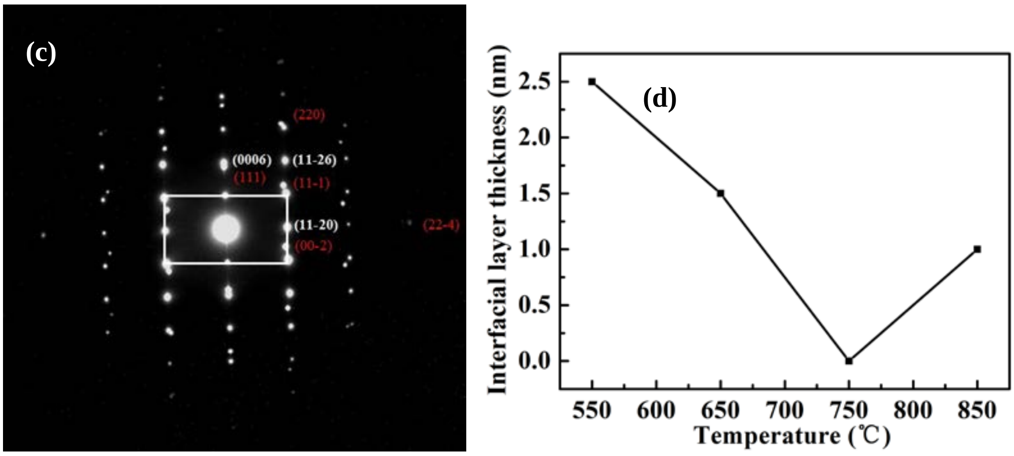


Fig. 7. Cross-sectional TEM images for Al epitaxial films grown on sapphire substrates. (a) High magnification TEM image for the interface between Al and sapphire grown at 750, and (b) 850 ºC. (c) SAED image for Al/Al_2O_3 grown at 750 ºC, where the spots marked in red correspond to planes of Al, and the spots marked in white correspond to planes of sapphire. (d) Temperature dependence of the interfacial layer thickness between Al and sapphire at temperatures ranging from 550 to 850 ºC.

4. Conclusions

In summary, 2-inch high-quality Al epitaxial films have been grown on sapphire substrates by MBE. The as-grown about 200-nm-thick Al epitaxial films are characterized by white-light interferometry, *in-situ* RHEED, FESEM, AFM, HRXRD, and HRTEM for surface morphologies and structural properties. These characterizations for 200-nm-thick Al epitaxial films grown at 750 ºC show homogeneous thickness distribution over the whole substrate, very flat surface with a RMS roughness of 0.6 nm, and high crystalline quality with an FWHM of 0.05°. There is no interfacial layer existing between Al epitaxial films and sapphire substrates at the growth temperature of 750 ºC. Furthermore, as the growth temperature increases from 550 to 850 ºC, the surface morphologies, crystalline qualities, and interfacial properties for 200-nm-thick Al epitaxial films improve dramatically at first and then decrease. Meanwhile, it also indicates an optimized result at the growth temperature of 750 ºC. We attribute this achievement mainly to two aspects. One is the very small lattice mismatch between Al epitaxial films and sapphire substrates, the other is the suitable growth temperature. The former is beneficial to the nucleation of Al epitaxial films and leads to the formation of few dislocations, and thereby enhances the growth of higher-quality Al epitaxial films. The latter provides enough energy for the migration of Al precursors on the substrate surface, enhances the growth process, and eventually leads to the growth of higher-quality Al epitaxial films. Although high-quality Al films have been grown on sapphire substrates by MBE, future work will be focused on the growth of high-quality oxide films and semiconductor films in turn on this high-quality Al epitaxial films, as well as preparation of the metal-oxide-semiconductor devices. Nevertheless, this work presents an effective approach for achieving large-size and high-quality Al epitaxial films for the future application of Al-based microelectronic devices.

Acknowledgements

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Table of contents

Title: Epitaxial growth and characterization of high-quality aluminum films on sapphire substrates by molecular beam epitaxy

Descriptions: 2-inch high-quality Al epitaxial films with sharp and abrupt Al/Al_2O_3 interfaces have been grown on sapphire substrates by molecular beam epitaxy with an in-plane alignment of Al[1-10]//Al_2O_3[1-100].