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Effect of Al evaporation temperature on the properties of Al films grown on sapphire substrates by molecular beam epitaxy

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Abstract: High-quality Al films with an in-plane epitaxial relationship of Al[1-10]//sapphire[1-100] have been epitaxially grown on sapphire substrates by molecular beam epitaxy. The as-grown about 200 nm-thick Al films with the Al evaporation temperature of 1100 ºC show high crystalline quality with the full-width at half-maximum of 180 arcsec, and very smooth surface with the root-mean-square roughness of 0.6 nm. There is no interfacial layer existing between Al and sapphire. Furthermore, the effect of Al evaporation temperature on the properties of the as-grown about 200 nm-thick Al films is studied in detail. This work of achieving high-quality Al films is of great importance for the fabrication of high-performance Al-based devices.

Keywords: epitaxial growth; thin films; X-ray technique; morphologies; structural

1. Introduction

Due to its superior properties, there is an increasing interest in the epitaxial growth of Al film[1-4] for the application of metal-oxide-semiconductor microelectronic devices and subwavelength surface plasmonic devices.[5-7] Recent studies find that the performance of these Al-based devices lies on the surface morphologies, crystalline qualities, and interfacial properties of Al films.[8-10] On one hand, the flat Al surface will reduce resistance of devices, and eventually benefit to obtaining high-efficiency devices. On the other hand, the high crystalline qualities of Al films mean that few dislocations are formed in the as-grown Al films, which helps to reduce the leakage current in devices. Furthermore, the high interfacial properties of Al/oxide hetero-interfaces are beneficial to the migration of electrons in devices. In this regard, the growth of high-quality Al films is of paramount importance for the fabrication of high-performance Al-based devices.[11-12] So far, there have been some reports about growing Al films. H. Liu et al. and H. Uchida et al. grew Al films on Si substrates by molecular beam epitaxy (MBE), and studied the Al surface morphologies and the corresponding epitaxial relationships.[6-7] However, due to large lattice mismatch between Al and Si, high-quality Al films on Si substrates are hard to be obtained. D. Medlin et al. used sapphire as substrate for the growth of Al films by the evaporation of Al from an effusion cell.[5] Thanks to the small lattice mismatch between Al and sapphire, the quality of Al films has been improved to some extent. However, the Al/sapphire hetero-interfaces are still not abrupt, and there is a ~1 nm-thick interfacial layer existing between Al films and sapphire substrates.[5] This may be ascribed to that the suitable growth condition for the Al films on sapphire substrates has not been deduced.

In this work, we epitaxially grow Al films on sapphire substrates by MBE with suitable Al evaporation temperature. On one hand, sapphire substrate shows a very small lattice of 3.9% with Al films, which is good for the nucleation of Al films during the initial growth. On the other hand, the suitable Al evaporation temperature in this work is beneficial to the migration of Al precursors. Both of these two aspects would lead to the growth of high-quality Al films.

Herein, we report on the growth of high-quality Al films with sharp and abrupt hetero-interfaces on sapphire substrates by MBE. The effect of Al evaporation temperature on the surface morphologies, crystalline qualities and interfacial properties of the as-grown about 200 nm-thick Al films is also studied in detail by various measurements, such as *in-situ* reflection high energy electron diffraction (RHEED), white-light interferometry, atomic force microscopy (AFM), high-resolution X-ray diffraction (HRXRD), and high-resolution transmission electron microscopy (HRTEM), respectively. It is found that the as-grown about 200 nm-thick Al films with the Al evaporation temperature of 1100 ºC show very smooth surface, high crystalline qualities, and sharp and abrupt hetero-interfaces. This work of growing high-quality Al films is of paramount importance for the application of Al-based devices.

2. Experimental

The as-received 2-inch sapphire substrates were taken a degassing treatment in an ultra-high vacuum (UHV) load-lock chamber with a background pressure of 1.0×10^{-8} Torr for 30 min, and then were transferred into the UHV MBE growth chamber with a background pressure of 2.0×10^{-10} Torr. Afterwards, the as-transferred sapphire substrates were annealed at 850 ºC for 60 min to remove the surface contaminations. During the epitaxial growth, high-purity (99.9999%) aluminum slugs with 3.175 mm diameter × 3.175 mm length bought from *Alfa Aesar* were used as the precursors of Al. Meanwhile, the Al evaporation temperature was set at range from 1000 to 1150 ºC in nitrogen atmosphere with the optimized nitrogen flow rate of 1 sccm. The rotation rate of the sapphire substrates was kept at 5 round-per-min to guarantee the growth of homogeneous thickness Al films at 750 ºC. As for the calculation of the growth rate, the Al films grown for 30 min at various temperatures ranging from 1000 to 1150 ºC were carried out. After the epitaxial growth, the surface morphologies, crystalline qualities, and interfacial properties of the as-grown about 200 nm-thick Al films grown were characterized by *in-situ* RHEED, white-light interferometry (Y-Wafer GS4-GaN-R-405), AFM (Bruker Dimension Edge, 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 kV, which gives a point to point resolution of 0.17 nm).

3. Results and discussion

During the epitaxial growth, *in-situ* RHEED observation is deployed to monitor the growth process. Fig. 1a is the sharp and streaky RHEED patterns for sapphire substrates after annealing process, which confirms the very smooth sapphire surface and is advantageous for the subsequent growth.[13-14] Fig.1b reveals the clear and streaky RHEED patterns for the as-grown about 200 nm-thick Al films grown at 750 ºC with the Al evaporation temperature of 1100 ºC, which proves that the single-crystalline Al films with smooth surfaces have been obtained. Meanwhile, we also find that the single-crystalline Al films on sapphire substrates can be obtained with various Al evaporation temperatures ranging from 1000 to 1150 ºC based on the RHEED observation. After carefully studying the RHEED patterns, an in-plane epitaxial relationship of Al[1-10]//sapphire[1-100] is obtained.[15-17] It is known that the *a* for sapphire is 0.4765 nm, and the corresponding length of $a_{sapphire}$/3[1-100] is 0.2751 nm; while the a for Al is 0.4040 nm, and the corresponding length of a_{Al}/2[1-10] is 0.2857 nm, as shown in Fig. 1c, respectively. Therefore, the lattice mismatch between sapphire[1-100] and Al[1-10] is calculated to be 3.9%.[18-19]

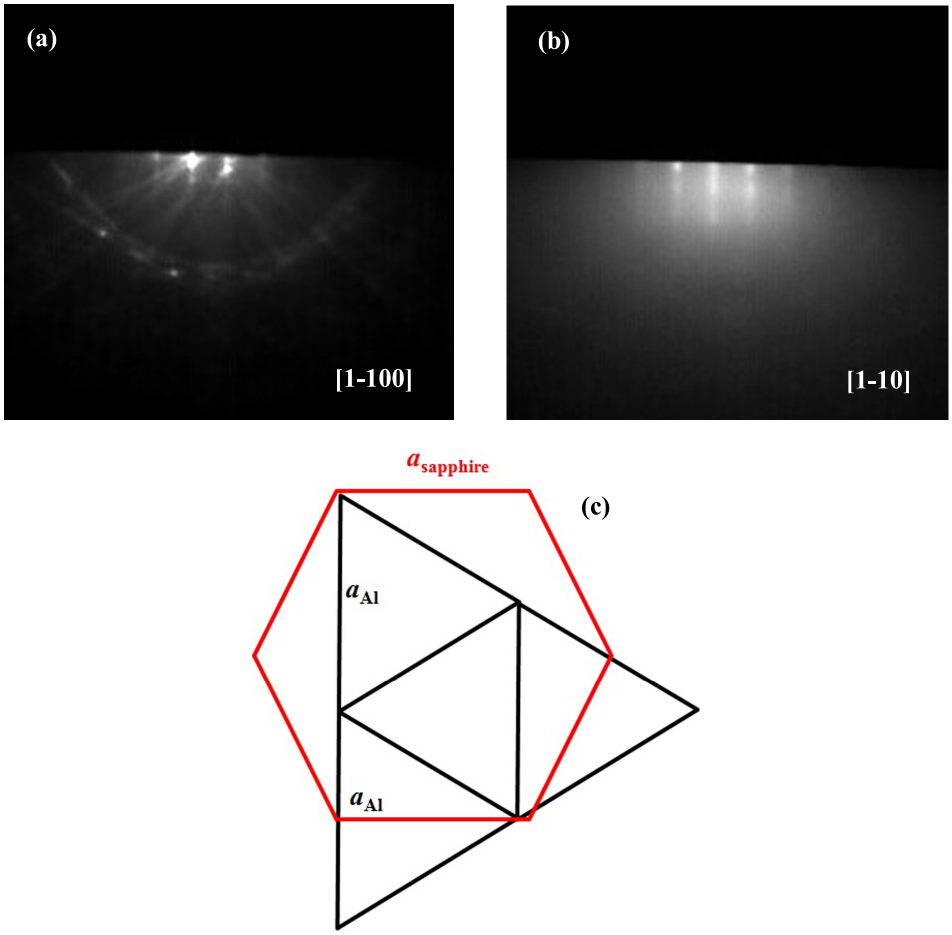

Fig. 1. RHEED patterns for as-annealed (a) sapphire substrates and (b) the as-grown ~200 nm-thick Al films grown with evaporation temperature of 1100 ºC. (c) The schematic diagram of lattice arrangement of Al films grown on sapphire substrates.

To calculate the growth rate of Al films grown with various Al evaporation temperatures, the white-light interferometry is deployed. Fig. 2a is a thickness distribution image of Al films grown on 2-inch sapphire substrates for 30 min with Al evaporation temperature of 1100 ^{o}C, and the thickness of the as-grown Al films is measured to be about 200 nm. Using the same method, the thickness for those Al films grown for 30 min with the Al evaporation temperature of 1000, 1050, and 1150 ^{o}C is measured to be about 159, 178, and 212 nm, respectively. Therefore, the average growth rate for Al films grown with the Al evaporation temperature of 1000, 1050, 1100, and 1150

°C is about 318, 356, 400, and 424 nm/h, respectively, Fig. 2b. In this case, we find that the growth rate for Al films grown on sapphire substrates is increased gradually with the Al evaporation temperature ranging from 1000 to 1150 °C.

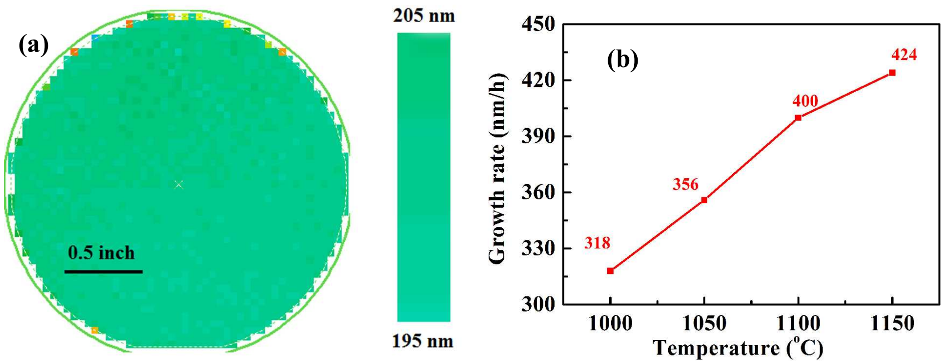

Fig. 2. (a) The thickness distribution of Al films on 2-inch sapphire substrates. (b) The growth rate of Al films grown with various Al evaporation temperatures ranging from 1000 to 1150 °C.

The Al evaporation temperature dependence of surface morphologies for the as-grown about 200 nm-thick Al films is studied by AFM. It can be clearly noted that the about 200 nm-thick Al films grown with Al evaporation temperature of 1000 ºC show very rough Al surface with the root-mean-square (RMS) surface roughness of 2.8 nm in Fig. 3a. If the Al evaporation temperature increases from 1000 to 1100 ºC, the surface morphologies are improved gradually. Actually, the about 200 nm-thick Al films grown with Al evaporation temperature of 1100 ºC show a very smooth surface with the RMS surface roughness of 0.6 nm, as shown in Fig. 3b. However, if the Al evaporation temperature is further increased, the surface morphologies for the about 200 nm-thick Al films become poorer with the RMS surface roughness of 2.1 nm, as shown in Fig. 3c. The possible reason can be attributed to the suitable Al evaporation temperature. If the Al evaporation temperature is too low, the flow rate for Al atoms will be low as well, which results in the low growth rate of Al films. During the growth, the low growth rate results in the poor surface due to the fact that the surface kinetics for the adatoms migration to their lowest energy crystal positions are not enough in this case. Furthermore, these adatoms are superimposed by additional adatoms, and many dislocation are formed in this case.[20-21] If the Al evaporation temperature is too high, the flow rate for Al atoms will be high as well, which would lead to the high growth rate of Al films. The high growth rate of Al films results in the rough surface due to the fact that the surface kinetics do not allow sufficient time for adatoms to move into their lowest energy crystal positions before being superimposed by additional adatoms.[22-23] Actually, many dislocations are also formed in this case.[22-23] Both of these two cases lead to the poor-quality of Al films. In this regard, the Al evaporation temperature of 1100 ºC seems to be the optimized temperature for the growth of high-quality Al films on sapphire substrates.

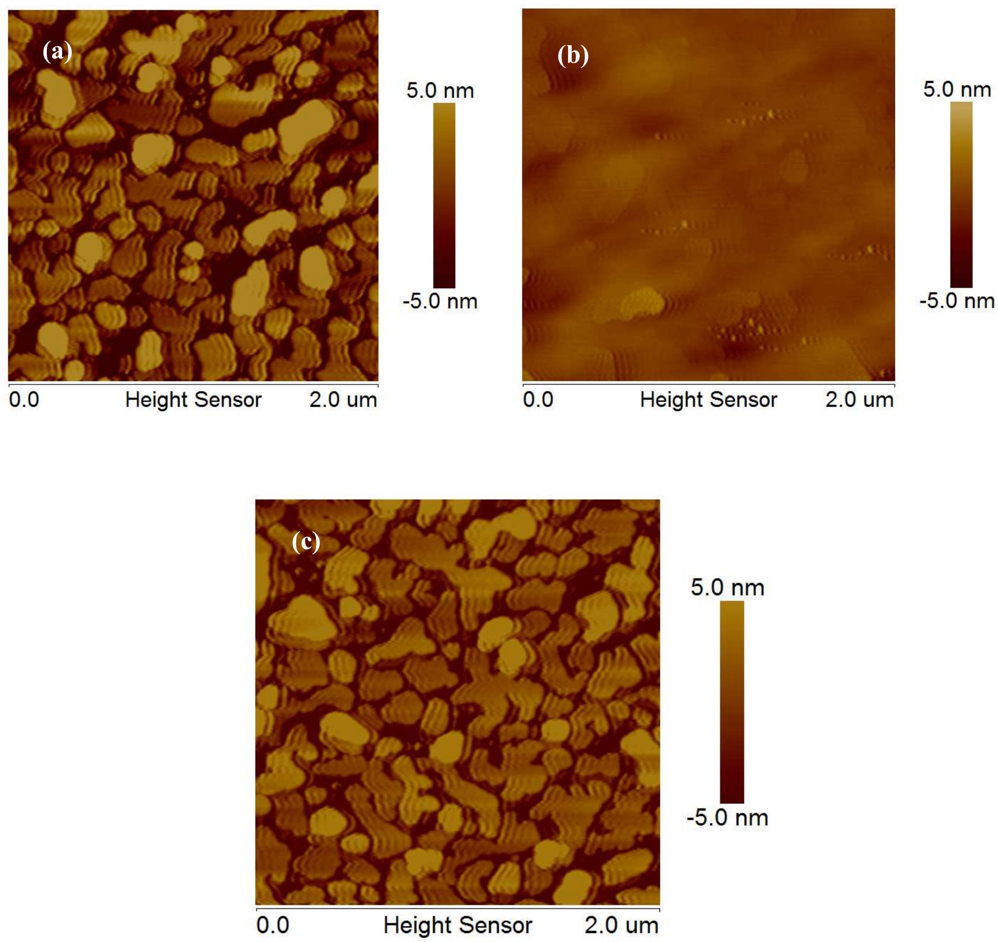

Fig. 3. AFM images for the as-grown about 200 nm-thick Al films grown on sapphire substrates with different Al evaporation temperatures of (a) 1000, (b) 1100, and (c) 1150 ºC, respectively.

XRD measurement is deployed to further study the structural properties of the as-grown about 200 nm-thick Al films. Fig. 4a is a typical XRD 2θ-ω scan for the about 200 nm-thick Al films grown with various Al evaporation temperatures. It can clearly find that when the Al evaporation temperature increases from 1000 to 1100 ºC, the peaks for Al(111) and Al(222) monotonously become sharp and narrow, which reveals the increase in crystalline quality.[11] However, when the is further increased to 1150 ºC, the peaks for Al(111) and Al(222) broaden, which confirms the decrease in crystalline quality. Furthermore, the out-of-plane epitaxial growth relationship of Al(111)//sapphire(0001) can be deduced from Fig. 4a. Fig. 4b shows the typical φ scans for Al(1-13) and sapphire(1-102), where the six-fold rotational peaks for Al(1-13) and three-fold rotational peaks for sapphire(1-102) can be identified. This result proves the in-plane epitaxial relationship of Al[1-10]//sapphire[1-100] between Al and sapphire,[24] which is well consistent with the RHEED observation. Based on these results, another in-plane epitaxial relationship of Al[11-2]//sapphire[11-20] between Al and sapphire can be obtained, as shown in Fig. 4c.

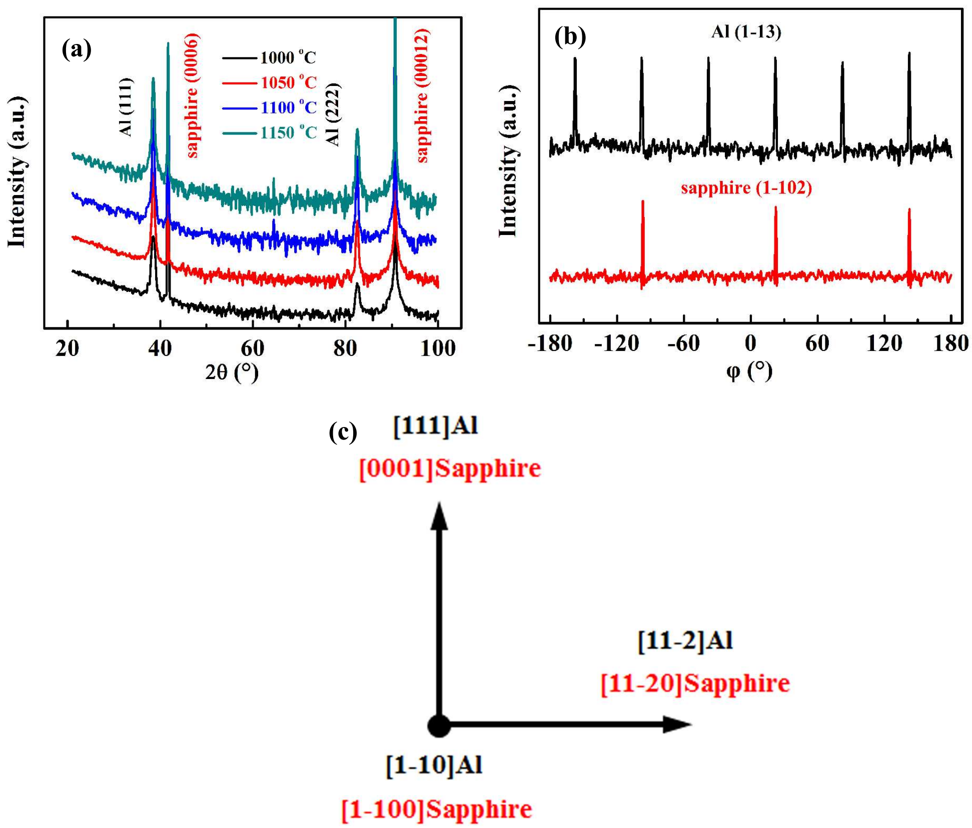

Fig. 4. XRD 2θ-ω scan for the as-grown about 200 nm-thick Al(111) films grown with various Al evaporation temperatures, and (b) φ scans for Al(1-13) and sapphire(1-102). (c) The epitaxial relationship between Al films and sapphire substrates for Al films grown on sapphire substrates.

The crystalline qualities of the as-grown about 200 nm-thick Al films are evaluated by X-ray rocking curves (XRCs). It is known that the full-width at half-maximum (FWHM) of XRC for Al films is related to the dislocation density in the films, and therefore the FWHM is widely used to evaluate the crystalline quality of as-grown films.[25-26] Fig. 5a is a XRC for the about 200 nm-thick Al films grown with the Al evaporation temperature of 1100 ºC, and the FWHM for Al(111) is as small as 180 arcsec, which is in striking contrast to that of Al films grown by Czochralski method with the FWHM of 1800 arcsec. We attribute this result to the very small lattice mismatch between Al and sapphire, which is good for the nucleation of Al films on the sapphire substrates during the initial growth and thereby is beneficial to the growth of high-quality Al films ultimately.[5, 11] Additionally, the Al evaporation temperature dependence of FWHM for Al(111) films is also studied, Fig. 5b. One can identify that when the Al evaporation temperature increases from 1000 to 1100 ºC, the FWHM for Al(111) films is greatly reduced from 1080 to 180 arcsec, which shows that the crystalline qualities of Al films are improved significantly. However, when the Al evaporation temperature is further increased to 1150 ºC, the FWHM for Al(111) films is broadened to be 288 arcsec, revealing the decline in crystalline quality.

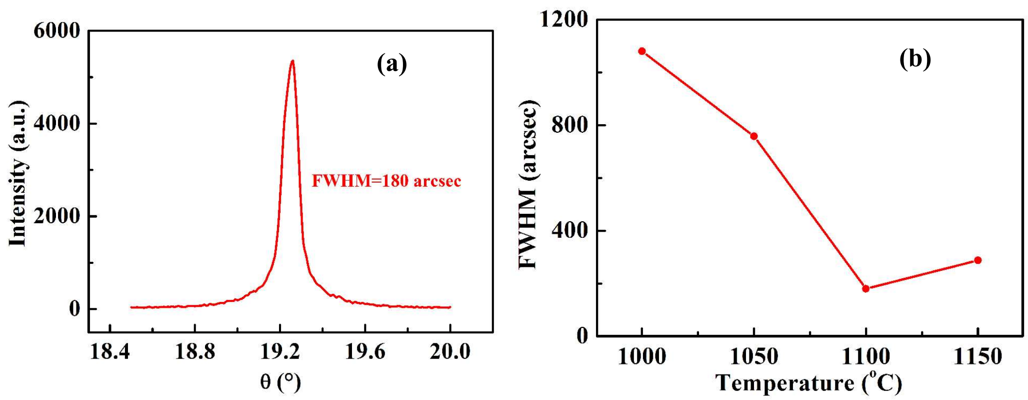

Fig. 5. (a) XRC for the about 200 nm-thick Al(111) films grown with the Al evaporation temperature of 1100 ºC and (b) The Al evaporation temperature dependence of FWHM for Al(111).

The interfacial properties of the as-grown about 200 nm-thick Al films grown on sapphire substrates are studied by cross-sectional TEM measurement. Fig. 6a is a low-magnification cross-sectional TEM image for 201 nm-thick Al films grown with Al evaporation temperature of 1100 ºC. Fig. 6b shows a high-magnification HRTEM image for the Al/sapphire hetero-interfaces grown with the Al evaporation temperature of 1100 ºC. There is no interfacial layer existing between Al and sapphire.[27-28] This result confirms the high interfacial properties of the as-grown Al films grown with the Al evaporation temperature of 1100 ºC. After careful study, another in-plane epitaxial relationship of Al[11-2]//sapphire[11-20] can be obtained.[29-31] Meanwhile, the Al evaporation temperature dependence of interfacial layer thickness for Al/sapphire hetero-interfaces is deduced, as shown in Fig. 6c. One can find that when the Al evaporation temperature is increased from 1000 to 1100 ºC, the interfacial layer thickness of Al/sapphire hetero-interfaces is decreased from 2.0 to 0 nm. However, when the Al evaporation temperature is further raised, the interfacial layer is increased to be 1.0 nm. We attributed this to the suitable Al evaporation temperature, which is beneficial to the migration of Al precursors on the sapphire substrates during initial growth and eventually leads to the sharp and abrupt hetero-interfaces. Meanwhile, the tendency of the interfacial layer thickness is well consistent with that of surface morphologies of Al films grown on sapphire substrates with various Al evaporation temperatures. In this regard, the Al evaporation temperature of 1100 ºC is the optimized temperature.

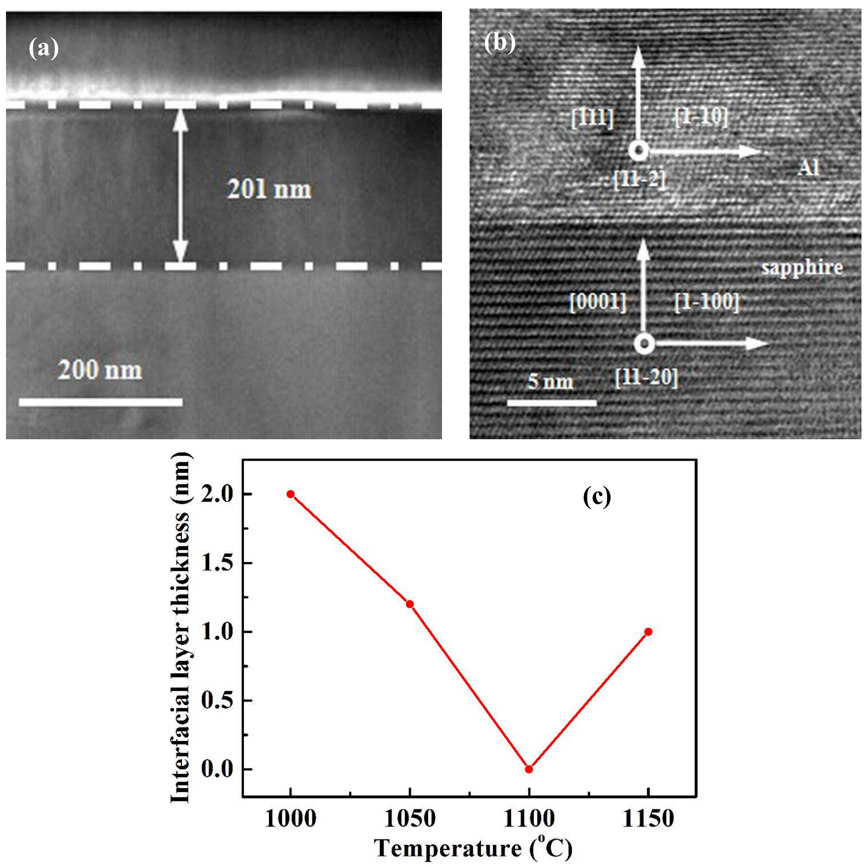

Fig. 6. (a) Low- and (b) high-magnification cross-sectional TEM images for Al/sapphire hetero-interfaces grown with Al evaporation temperature of 1100 ºC. (c) The Al evaporation

temperature dependence of interfacial layer thickness.

4. Conclusions

In summary, the about 200 nm-thick Al films have been grown on sapphire substrates with various Al evaporation temperatures. The Al evaporation temperature dependence of crystalline qualities, surface morphologies, and interfacial properties for the as-grown about 200 nm-thick Al films grown on sapphire substrates are carefully studied. It is found that the crystalline qualities, surface morphologies, and interfacial properties for as-grown about 200 nm-thick Al films are first improved and then decreased as the Al evaporation temperature increases from 1000 to 1150 ºC, and show the optimized values at Al evaporation temperature of 1100 ºC. We ascribe these results to two aspects. One is the small lattice mismatch between Al and sapphire, and the other is the suitable Al evaporation temperature. The former is advantageous for the nucleation of Al films on sapphire substrates, and is beneficial to the growth of high-quality Al films. The latter is good for the coherence of Al precursors. If the Al evaporation temperature is too low, the flow rate for Al atoms will be low as well, which results in the low growth rate of Al films. During the growth, the low growth rate results in the poor surface due to the fact that the surface kinetics for the adatoms migration to their lowest energy crystal positions are not enough in this case. Furthermore, these adatoms are superimposed by additional adatoms, and many dislocation are formed in this case. If the Al evaporation temperature is too high, the flow rate for Al atoms will be high as well, which would lead to the high growth rate of Al films. The high growth rate of Al films results in the rough surface due to the fact that the surface kinetics do not allow sufficient time for adatoms to move into their lowest energy crystal position before being superimposed by additional adatoms. Actually, many dislocations are also formed in this case. Both of these two cases lead to the poor-quality Al films. This work of obtaining high-quality Al films is of great importance for the fabrication of high-performance Al-based devices.

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