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Influence of substrate biasing on the growth of c-axis oriented AlN thin films by RF reactive sputtering in pure nitrogen

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We report on the investigation of the influence of deposition conditions on structural, morphological and optical properties of AlN thin films deposited on sapphire (Al_2O_3) substrates by radio-frequency (RF) reactive sputtering. The deposition parameters studied are RF power, substrate temperature and substrate bias, while using pure nitrogen as reactive gas. The effect of such deposition parameters on AlN film properties are analyzed by different characterization methods as high resolution X-ray diffraction (HRXRD), field emission scanning electron microscopy (FESEM) and linear optical transmission. AlN thin films with a full-width at half-maximum (FWHM) of the rocking curve obtained for the (0002) diffraction peak of 1.2º are achieved under optimized conditions. The time resolved evolution of the self and externally-induced biasing of the substrate during deposition process is monitored and analyzed in terms of the rate of atomic species incorporation into the layer. The bias-induced change of the atomic incorporation leads to an enhancement in the structural quality of the layer and an increase of the deposition rate.

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1 Introduction Aluminum nitride (AlN) has received considerable interest due to its important properties such as a wide direct band gap (~ 6.2 eV) [1], high thermal conductivity, high acoustic velocity and piezoelectricity. These properties make AlN material appropriated for a wide variety of applications in optoelectronics (UV spectral range), in surface acoustic wave devices (SAW) and in microelectromechanical systems (MEMS) [2]. In most of these applications, a highly quality AlN film with smooth surface is required.

Many techniques, such as metalorganic vapour phase epitaxy (MOVPE) and molecular beam epitaxy (MBE) are used to obtain AlN thin films on different substrates. The growth of AlN layers usually requires the use of high substrate temperature, which could deteriorate the substrate quality and thus the AlN properties during the growth process [3]. Hence, among the growth techniques generally used for AlN, RF reactive magnetron sputtering emerges as a low-cost solution which allows deposition in both rigid and flexible substrates and in a wide range of temperatures.

In this work, we present the successful growth of AlN layers on sapphire by RF reactive sputtering under pure nitrogen atmosphere. Firstly, we discuss the effect of various deposition parameters, such as RF power and substrate temperature on crystalline structure, surface morphology and optical properties of AlN films. Besides, the effect of substrate biasing during deposition on the thickness, strain, grain size and apparent bandgap energy is analyzed.

2 Experimental The samples studied in this work consist of AlN layers deposited on 500 µm-thick c-axis oriented sapphire substrates using an RF sputtering system equipped with a 2'' confocal magnetron cathode (AJA International, ATC ORION-3-HV). A high-purity Al disk (5N) was used as the Al target whereas the reactive gas was pure N_2 (6N). The target-substrate distance was kept at

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10.5 cm for all samples. A thermocouple placed in contact with the substrate holder allowed the monitorization of substrate temperature during deposition processes. Prior to growth, the substrates were cleaned in organic solvents and blown dry with nitrogen. Then, they were loaded in the sputtering system, which is pumped down to a base pressure of ~ 10^{-7} mBar. Substrates were outgassed in the deposition chamber for 30 min at a substrate temperature 100 ºC above the growth temperature.

The structural characterization of the deposited AlN films was performed by high resolution X-ray diffraction (HRXRD). A Bruker D8-4C Advance X-ray diffractometer was used to analyze both the crystal orientation and the structural quality of the layers by 2θ/ω and rocking curve around (0002) diffraction peak measurements, respectively. The layer grain size (G) is estimated by means of the Sherrer formula

$$G = \frac{0.9 \cdot \lambda}{\Delta 2\theta} \cdot \cos\theta \qquad (1)$$

in which $\Delta 2\theta$ is the FWHM of the AlN(0002) diffraction peak in the 2θ/ω measurement, λ is the Cu $K\alpha_1$ wavelength and θ the diffraction angle corresponding to the (0002) orientation. XRD characterization was also used to determine the layer thickness by performing grazing incidence X-ray reflectivity (XRR) measurements.

The morphological analysis was carried out by FESEM technique using a FEI Nova Nanosem 230 microscope with VCD detector.

The linear optical properties were studied from transmission measurements in the 180-2500 nm spectral range, obtained under normal incidence and at room temperature using a UV/VIS/NIR Perkin-Elmer Lambda 1050 scanning spectrophotometer. The AlN bandgap energy was calculated by considering a sigmoidal fitting of the optical absorption obtained from transmission measurements.

3 Results and discussion

The sputtering parameters studied in this work for the deposition of AlN layers are summarized in Table 1.

Table 1 AlN sputtering parameters

Parameter	Value
Deposition pressure	3.5 mTorr
Reactive gas, N_2	100 %
RF power	100, 125, 150, 175 W
Substrate temperature	350, 400, 450, 500 ºC
Substrate bias	Floating, +5, -5, -25 V
Deposition time	2 h

As it is shown in Table 1, this work is focused on the investigation of the effect of three deposition parameters, the RF power, the substrate temperature and the substrate bias on the AlN layer quality. For all the experiments the deposition working pressure was fixed to 3.5 mTorr, using pure nitrogen as reactive gas. Both conditions were taken for previous results obtained in the growth optimization process of InN layers in the same deposition system [4].

3.1. Effect of RF power

The effect of RF power on the AlN quality was studied through a set of samples deposited at RF powers ranged from 100 to 175 W (see Table 1). For this set of samples the substrate temperature are kept fixed to 400 ºC, while the samples are not externally biased (floating substrate configuration).

Figure 1 shows a representative 2θ/ω scan around the (0002) x-ray reflection peak of the AlN. It must be pointed out that all the samples deposited to investigate the effect of RF power present the reflection peak associated with (0002)-oriented AlN without parasitic crystallographic orientations.

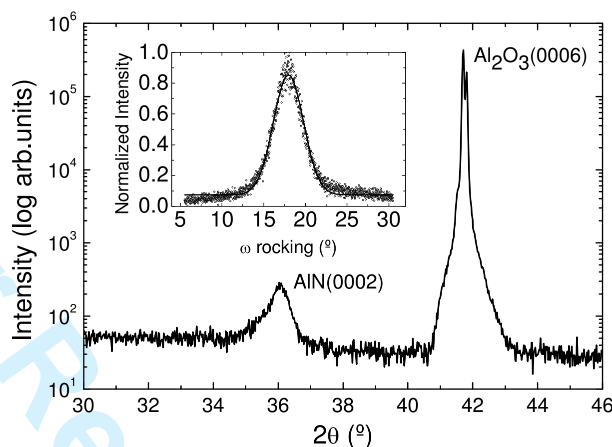

Figure 1 XRD 2θ/ω scan of AlN films deposited on Al_2O_3 substrates at 100% N_2, 3.5 mTorr, 100 W, 400 ºC and floating substrate. Inset: XRD rocking curve of the AlN(0002) reflection peak fitted to a gaussian curve.

The estimated c-lattice parameter for AlN films under the previous deposition conditions is about 4.983 Å, which is close to the corresponding one to fully relaxed AlN films, of 4.982 Å [5].

Figure 2 shows the ω-scan XRD measurements of the set of samples deposited for this study. The FWHM of the rocking curve obtained for the AlN(0002) diffraction peak is decreased from 4.26º to 2.18º when increasing the deposition RF power from 100 to 150 W. At the same time, an increase of the growth rate from 38 to 45 nm/h is estimated using FESEM measurements when increasing the RF power from 100 to 150 W, respectively. This improvement of crystal quality and growth rate with the RF power is in agreement with results presented by other authors [6][7]. It is worth saying that no deposition was detected for RF power below 100 W. Further increase of this deposition parameter up to 175 W leads to a degradation of the layer crystalline quality. For this sample a FWHM of the (0002)

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diffraction peak in the ω-scan of 3.8º is obtained. This deterioration of crystal quality is attributed to surface damage generated by the high energy Al ions during deposition process.

At the same time, the estimated grain size of the studied AlN films follows a similar trend that the FWHM of the ω-scan, with values of 14, 19, 26 and 22 nm for RF powers of 100, 125, 150 and 175 W, respectively. Taking into account the previous results, the RF power was fixed to 150 W for the following experiments.

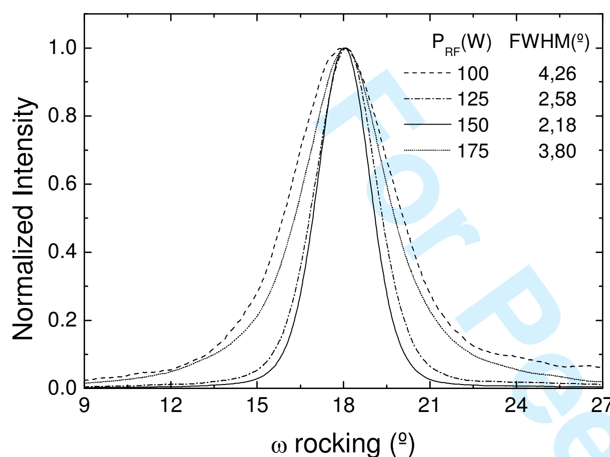

Figure 2 Normalized XRD ω scan of AlN films deposited on Al_2O_3 substrates at 100% N_2, 3.5 mTorr, 400 ºC and floating substrate as a function of the RF power.

3.2 Effect of substrate temperature For this study, the substrate temperature was changed in the range of 350 ºC to 500 ºC. 2θ/ω XRD patterns of the samples show that changes in substrate temperature do not have an influence on the preferred orientation of the layers. Regarding the structural quality, a slight decrease of the FWHM of the rocking curve is achieved from 2.18º to 2.07º at a temperature of 450 ºC. On the other hand, layers deposited at 350 and 500 ºC show an increase of the FWHM to 8.77º and 5.78º, respectively. The deterioration in the structural quality of the c-axis orientation growth for AlN films at higher temperature can be attributed to resputtering of atoms from the substrate [8]. Considering all these results, the optimized temperature to reach high-quality c-axis oriented AlN thin films, under the previously fixed deposition conditions, is 450 ºC.

This optimized layer deposited at 100% N_2, 3.5 mTorr, 150 W, 450 ºC and floating substrate presents a c-axis lattice parameter of 4.973 Å. Thus, it shows a residual tensile strain (ε_{zz}) of $1.7 \cdot 10^{-3}$. The grain size is about 26 nm with a growth rate of ~50 nm/h.

Figure 3 shows optical transmission measurements of the AlN layer deposited under optimized growth conditions. The absorption coefficient of the sample was derived from linear optical transmission (T) measurements using the relation $\alpha(\lambda) \propto -\ln(T)$, which neglects the losses introduced by interfaces reflection and optical scattering. The obtained absorption was modeled using a sigmoidal approximation that allows the bandgap energy estimation as the intersection of the linear fitting to α^2 (E) with the X-axis.

Figure 3 shows the above described fitting for the optimized sample, which leads to a bandgap estimation of 6.0 eV, in good agreement with the obtained one in sputtered AlN samples [9].

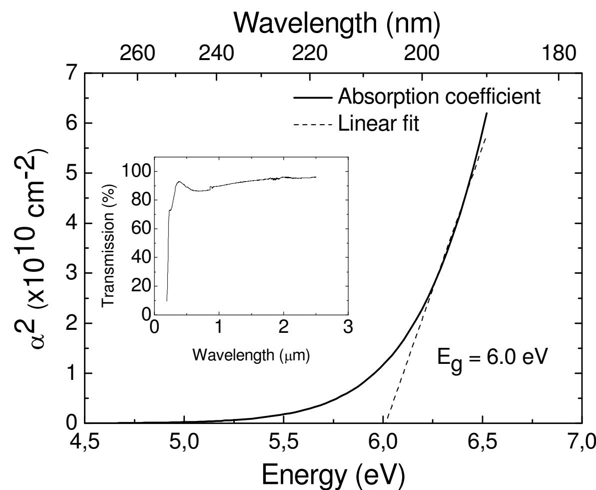

Figure 3 Linear fit of the experimental square absorption coefficient of AlN layer deposited onto c-axis sapphire substrates at 100% N_2, 3.5 mTorr, 150 W, 450 ºC and floating substrate. Inset: Optical transmission spectrum of the same sample in the 180-2500 nm spectral range.

3.3 Effect of substrate biasing Once AlN films are optimized in terms of RF power and substrate temperature, the third deposition parameter studied was the substrate biasing, which was changed between +5 to -25 V (see Table 1).

For this experiment, a DC voltage source is connected to the substrate holder while the current intensity is measured as a function of deposition time. In the studied electrical configuration the deposition chamber is electrically grounded.

For all the biased samples, the measured time-dependent evolution of the current intensity follows a similar behavior than one showed for a capacitor charging process. Namely, after an exponential growth, the current intensity saturates to a value which increases with the negative externally-induced bias (see Figure 4), changing from 0.4 to 2.5 mA for substrate bias of +5 to -25 V, respectively. This evolution and the dependence of the saturation intensity with the substrate bias is explained in terms of screening of the externally induced electric field in the sample from the impinging charged species present in the plasma during deposition.

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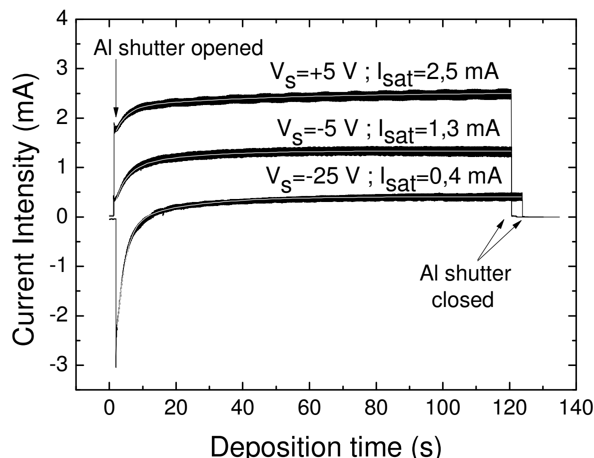

Figure 4 Real-time current intensity evolutions during depositions with external-induced bias as +5 V, -5 V and -25 V.

Figure 5 shows the ω-scan XRD measurements of layers deposited under optimized deposition conditions and different substrate bias. A decrease of the FWHM values of the rocking curve is observed when increasing the negative substrate bias. This enhancement in the structural quality of the layers is related to the higher energy of the Al ions which reach the substrate under biasing conditions, enabling the migration of the adatoms at the growing surface. As it has been referred by other authors, the (0002) plane is favorable to grow when the atomic species have a high energy, as it involves the two types of bonds found in wurtzite structure [10].

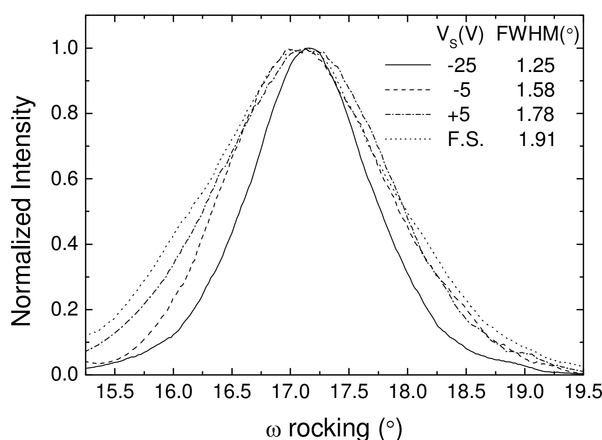

Figure 5 ω-scan XRD measurements of AlN films deposited on Al_2O_3 substrates at 100% N_2, 3.5 mTorr, 150 W and 450 ºC as function of the substrate bias applied, compared to the unbiased layer (in floating substrate configuration, F.S.).

Figure 6 shows the evolution of the grain size and the c-axis lattice parameter depending on the applied substrate bias during AlN deposition. The highest grain size is obtained for substrate bias of -25 V, with a grain size of 52 nm, which corresponds to a FWHM of the (0002) diffraction peaks obtained in 2θ/ω scan of 1.2º, well below the obtained one of 1.9º for samples grown without biasing the substrate.

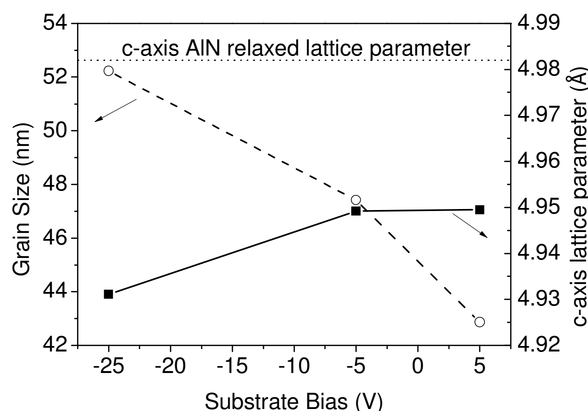

Figure 6 Dependence of grain size and c-axis lattice parameter on substrate bias for AlN layers deposited on sapphire at 100% N_2, 3.5 mTorr, 150 W and 450 ºC.

The c-axis lattice parameter decreases with the negative substrate biasing from 4.949 to 4.931 Å (see Figure 6), which corresponds to tensile strain (ε_{zz}) states ranging from 6.6×10^{-3} to 1.0×10^{-2}, respectively. This slightly increase in the residual strain in the layers is attributed to the increase of the grain size.

The layer thickness measured by XRR in grazing incidence condition is about 122 nm, hence, the growth rate increases in almost 20% compared to the optimized unbiased layers, reaching a deposition rate of about 61 nm/h. Figure 7 show a FESEM image of AlN layer deposited under optimized deposition conditions with substrate bias, showing a compact morphology of the layer.

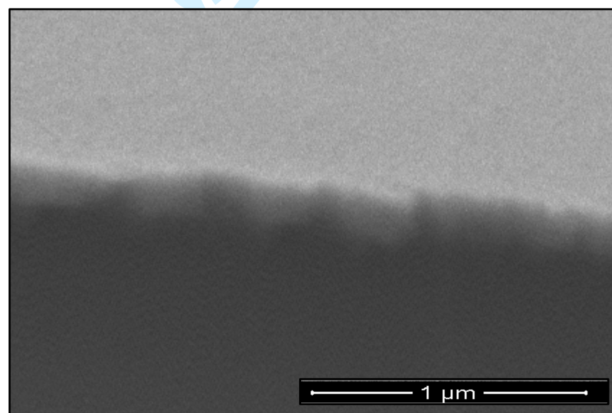

Figure 7 FESEM image of AlN layer deposited directly on c-axis sapphire biased substrate at -25 V and in a pure nitrogen atmosphere. The sputtering pressure, RF power and substrate temperature were 3.5 mTorr, 150 W and 450 ºC, respectively.

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Regarding the optical properties of the optimized AlN layers, a decrease of the estimated bandgap to 5.9 eV is observed for biased samples, it being attributed to an enhanced incorporation of impurities.

3 Summary The influence of deposition conditions such as RF power, substrate temperature and substrate bias on morphological, structural and optical properties on AlN films deposited on (0006)-oriented Al_2O_3 in pure nitrogen by RF reactive sputtering were analyzed. AlN films with a (0002) preferred growth orientation were deposited in optimized sputtering conditions without substrate bias. For the analyzed conditions, the structural quality of the samples deteriorates for RF powers and deposition temperatures above 150 W and 450 ºC, respectively.

AlN layers deposited biasing the substrate show an enhancement in crystalline quality on (0002) growth orientation. This result is attributed to an increase of the energy of the ions reaching the substrate due to the external biasing. Furthermore, for biased samples, the deposition rate increases while both grain size and residual tensile strain are enhanced.

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