ABSTRACT

Aluminium nitride is a promising coating material for many applications, due to its good physical and mechanical properties. Hard transparent Aluminium nitride thin films have been deposited by bipolar reactive pulsed magnetron sputtering (PMS). With its inherent advantages of energetic particle bombardment of the growing film and minimal arcing tendency even in dielectric layer deposition processes, the technology allows deposition of high quality films. Layers have been deposited with a rate of 53 nm/min on moving substrates in a batch coating device. Characterization of the process was performed in both a fixed flow mode and a current controlled mode at a working pressure of 0.2 Pa. Based on the characteristic curves obtained; different working points have been selected for the deposition of 1.5 µm thick AlN layers on Si-Wafers and floatglass. For film deposition, the reactive sputter process was operated with a closed-loop feedback control to maintain stable operating conditions during the coating process. The Influence of varying the working points on the mechanical and the physical properties of the layers have been studied. All films are stoichiometric and highly transparent. Mechanical properties have been determined on coated Si-Wafers. The films exhibit hardness up to 26 GPa and elastic modulus up to 336 GPa and these properties do not vary significantly with varying working point. In contrast, the residual stress can be tailored from +0.5 GPa tensile to –0.5 GPa compressive stress by appropriate choice of deposition conditions.

KEYWORDS
Reactive deposition; Pulsed sputtering; characteristic curves; AlN; Film properties;
1 INTRODUCTION

Aluminium nitride (AlN) is one of the III–V compound semiconductors that exhibits a close-packed wurtzite crystalline structure. It is a promising coating material in electronic applications, such as transducers in surface acoustic wave (SAW) devices and in microwave bulk acoustic wave (BAW) delay lines, insulation or passivation layers in integrated circuits, etc., because of its high longitudinal acoustic wave velocity, good piezoelectric and electric properties, relatively good thermal conductivity, and easy processing [1-4]. Also, AlN is an interesting optical material due to its wide energy band gap (6.3 eV) and relatively high refractive index, n>2.1, in the visible range [5]. Using AlN as a buffer layer is a very promising way to grow high-quality GaN on Si. The lattice and thermal mismatch between AlN and GaN are much smaller than those between Si and GaN, and AlN has good wetting properties on Si [6, 7]. With the coating of an AlN film, the surface hardness, corrosion resistance and the oxidation resistance of both steel [8] and magnesium alloys [9] can be increased significantly.

The growth of AlN films has been performed by various techniques such as reactive ion beam deposition, reactive evaporation, chemical vapour deposition and molecular beam epitaxy. Among all these techniques, either the growth rates are low or the substrate temperatures during the growth are high and incompatible with other processing steps in device fabrications. Reactive sputtering technique, however, offers the possibility of depositing high quality films at relatively low growth temperature. Recently, it has been demonstrated that highly c-axis oriented wurtzite AlN films can be obtained at a substrate temperature as low as 100 °C using the reactive sputtering technique [10]. In addition, PVD has more advantages over other deposition technologies such as Plasma Assisted Nitriding PAN, since it produces smoother, more uniform layers with similar thickness in shorter processing times [11].

For more than two decades, reactive sputtering of thin films has been intensively investigated because the sputtering of metallic targets in the presence of reactive gas makes it possible to easily form compound films, such as nitrides, oxides, carbides or their combinations. The reactive sputtering process can be divided into three modes according to the amount of reactive gas used in the film deposition: (a) metallic, (b) transition and (c) reactive. A typical characteristic of the reactive magnetron sputtering is a low deposition rate of compounds produced in the reactive mode compared to that of the pure metallic or alloyed films produced in the metallic mode. The ratio between the deposition rate in the metallic mode to that in the reactive mode is relatively small (3–4) for nitrides but achieves high (10–15) values for oxides [12].

The decrease in the deposition rate of films sputtered in the reactive mode is due to a reaction of the reactive gas with the surface of the sputtered target and its conversion to a compound. This decrease in deposition rate depends on the sputter coefficient (sputtering yield) of the compound that forms on the target [13]. When the reaction product is electrically insulating, two further problems occur: (1) non-sputtered surfaces of the target (very significant in planar magnetrons) are covered by thick dielectric layers, which are charged up and cause arcing when the charge achieves a threshold value; and (2) the efficiency of the anode of the magnetron decreases due to its progressive coverage with a dielectric layer cover. The first problem can be partly diminished by polishing these non-sputtered surfaces before starting the deposition process. The second problem is eliminated by using either a hidden anode, or by operating two magnetrons in a bipolar pulsed mode, i.e. bipolar dual magnetron sputtering (BP-DMS), Schiller et al. [14].
In BP-DMS, two targets are used as anode and cathode, respectively. During the cathode phase, the target is sputter-cleaned, hence ensuring a metallic surface during the anode phase and a stable long-term operation. At high-enough frequencies (25–50 kHz), possible electron charging of insulating layers will be minimised, thus limiting the troublesome phenomenon of arcing [15,16,18].

There have been extensive studies carried out for investigating the morphological, optical, and electrical properties of AlN thin films [1-5,19-24]. However, the published data concerning their mechanical properties is not as extensive [24,25]. In this paper, a study for the reactive Al-sputter process in Ar-N₂-gas mixture is presented. High quality AlN films were deposited with variation in process parameters. Dependence of both the deposition rate, film morphology, residual stresses and the mechanical properties of these films on growth conditions such as substrate temperature and the working point is investigated.

2 EXPERIMENTAL

Experiments have been carried out in a production scale batch coating device of volume 1 m³. The cuboid-shaped stainless steel recipient is evacuated with a diffusion pump down to a base pressure of 10⁻³ Pa (10⁻⁵ mbar). A rotary substrate holder allows the rotation of substrates about, up to, three parallel vertical axes (three-fold rotation), providing the possibility to coat 3D-shaped substrates uniformly on a peripheral surface. The device is equipped with a central heater. Two dual magnetron systems (DMS) are installed, each powered by a separate pulse voltage supply (MAGPULS Q-1000/60/500 BP). An additional power supply can be used for plasma pre-treatment as well as for substrate bias, either DC or pulsed. In order to deposit AlN films, two pure aluminium targets (4N purity) were installed to one of the DMSs. Target dimensions are 512 x 128 mm².

In most cases, AlN deposition was performed at a total pressure of approximately 0.21 Pa (2.1 µbar): a predetermined flow of 200 sccm Ar was fixed during the deposition processes, accompanied by a closed-loop-controlled nitrogen flow. During all deposition runs, the substrates were kept at floating potential. The substrate temperature was ranging between 300°C and 350°C.

1.4…1.6 µm thick layers were deposited on Si-wafers with (111) orientation (76.2 mm in diameter and 375 µm thick). Film thickness was measured using the standard CALOTEST®. Mechanical properties of the deposited films were measured using a Nano-Indentor XP® from MTS. The residual stresses of the coatings were calculated from curvature measurement obtained using a Tencor P15LS apparatus. Film morphology was investigated by a Scanning Electron Microscope DSM 962 (Zeiss).

2.1 Characteristic Curves

The process characteristic curve of AlN deposition process was recorded in both fixed reactive gas flow rate mode, and current controlled mode. In both cases a predetermined argon gas flow rate of 200 sccm was fixed. A bipolar sputter voltage of amplitude 800 V is applied between the two magnetrons of a DMS pair. Pulse on- and off-times have been chosen to (τ²ₘ₁ on: τ²ₘ₁ off: τ²ₘ₂ on: τ²ₘ₂ off) = (10:10:10:8) µs to accommodate slightly asymmetric behaviour of the magnetrons.
2.1.1 Fixed flow mode
In order to record the characteristic curve for the AlN deposition process in a fixed flow mode, the reactive gas flow rate was manually controlled. Gradually, the value for the nitrogen gas flow rate was increased up to 85 sccm. After some minutes from setting the value of the flow rate, the other process parameters were recorded, including: the DC current of the process, the optical emission values for each magnetron source, the power dissipated in the cooling water of each magnetron, temperature and pressure.

The experimental process curve for both the DC current and the optical emission vs. the supply flow rate of the reactive gas in a fixed flow mode of operation is shown in Fig. 1. It can be observed that the increase in the DC current with increasing the nitrogen flow is small in the more metallic region, followed by a steep increase in the transition region, and finally a saturation level is reached in the reactive region. No broad hysteresis is observed, when the reactive gas flow rate is decreased. However, a more or less pronounced jump in the DC current and optical emission values upon increasing the nitrogen flow rate from 65 to 67 sccm and slightly different values of these quantities upon decreasing the reactive gas flow again indicate a very narrow hysteresis for the chosen experimental conditions.

![Fig. 1: Fixed flow characteristic curves](image)

Similar behaviour is observed for the values of the optical emission for both magnetrons. It has to be mentioned that the values for the optical emission don’t have any quantitative significance, since they depend on the sensitivity of the optical emission detector used for each magnetron. Nevertheless, they give a good qualitative representation for the characteristic behaviour of the deposition process with respect to each magnetron.

2.1.2 Current controlled mode
In current controlled mode of operation, the nitrogen flow rate is regulated by a closed-loop process control. In order to record a process characteristic curve in a current controlled mode, we gradually changed the DC current setpoint for the controller, and at each point we recorded the process parameters previously mentioned. Fig. 2 shows the change in the value of the reactive gas flow rate with increasing the current setpoint, and the effect of this change on the optical emission values for each magnetron.

![Fig. 2: Current controlled characteristic curves](image)
Fluctuations were noticed in the three current setpoints of 23, 24 & 25 Amperes. The process does not display a pronounced transition region of negative slope, but steep characteristics in a narrow range of reactive gas flow rate around 60 sccm. The behaviour is consistent with the observations in the fixed flow mode. However, process control allows establishing any current setpoint, also in the steep transition region.

2.2 Film Depositions
Deposition processes were run in a current controlled mode. The current setpoint was changed from 20 A down to 14 A in steps of 2 A in consecutive deposition processes. Moreover, the effect of additional substrate heating was investigated. The following parameters were kept constant in all deposition runs: (a) the deposition time, \( t = 30 \text{ min} \), (b) the deposition pressure, \( p = 0.2 \text{ Pa} \), (c) Argon flow rate is fixed at 200 sccm Ar, (d) Both magnetrons are connected to power supply MagPuls, 800 V, and (e) Pulse times \((\tau_{\text{M1 off}}, \tau_{\text{M1 on}}, \tau_{\text{M2 off}}, \tau_{\text{M2 on}})\) are (10:10:10:8) \(\mu\text{s}\). Table 1 lists the deposition runs carried out and their respective process parameters.

<table>
<thead>
<tr>
<th>Depositio\n run</th>
<th>Current setpoint (A)</th>
<th>External heating</th>
<th>Approximate substrate temperature (°C)</th>
<th>Argon flow rate (sccm)</th>
<th>Average nitrogen flow rate (sccm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20</td>
<td>Yes</td>
<td>340</td>
<td>200</td>
<td>58</td>
</tr>
<tr>
<td>2</td>
<td>18</td>
<td>Yes</td>
<td>330</td>
<td>200</td>
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<td>5</td>
<td>16</td>
<td>No</td>
<td>225</td>
<td>200</td>
<td>57</td>
</tr>
</tbody>
</table>

The time behaviour of both the voltage and the current obtained at the output of the pulse generator is plotted in Fig. 3 (This figure represents the pulse time behaviour for the deposition process having a current setpoint of 20 A). In Fig 3(a), the potential of one magnetron source with respect to ground is plotted during the course of 200 \(\mu\text{s}\). Each sputtering phase of 10 microseconds duration is followed by a phase in which the insulating layers are discharged. In that way, arcing is prevented in a highly efficient manner. Moreover, the sputter process cleans the targets and therefore insures the stable long-term anode function.
of each target. During the on-time of this magnetron, the average value of voltage is about -600…-800 V (with respect to ground). However, it is about 0…200 V during the subsequent anodic phase. Fig 3(b) shows the time behaviour of electric current at the output of the pulse generator for both magnetrons. The current profile of Magnetron 1 is plotted in the lower half of the screen of the oscilloscope. While the current profile of Magnetron 2 is plotted in the upper part. In principal, the current increase during the on-phase of a magnetron source is a linear increase, i.e. a triangular-shaped profile. Nevertheless, some high frequency bursts of high amplitude peaks (higher than 100 A) can be observed. The occurrence of these HF bursts depends on the reactivity of the process, i.e. the chosen working point.

3 INVESTIGATION RESULTS & DISCUSSION

The investigations carried out on the deposited films included measuring the film thickness, residual stresses and mechanical properties. Moreover, Scanning Electron Microscope images were obtained in an attempt to correlate the films structure to its properties. In addition, the effect of additional substrate heating was studied. All films deposited are stoichiometric and highly transparent.

3.1 Deposition Rate

Fig. 4 shows the variation in both, film thickness and deposition rate resulting from changing the DC current set-point for the deposition process. Moreover, the effect of heating is illustrated. The thickness varied between 1.4 to 1.6 µm due to a variation in deposition rate.
from 46.7 up to 53.3 nm/min. The deposition rate increases with increasing current setpoint, which also means increased power input, up to a value of 18 A. However, the deposition rate was constant from 18 A to 20 A. Relating the deposition rate to the power dissipated in the cooling water of the magnetrons, which is a measure for the sputtering power, a decrease in rate per unit power is found with increasing working point or increasing reactivity of the process.

3.2 Film Stresses
The stress behaviour of films is very important in all applications of thin films with respect to durability and hence use. The presence of mechanical stresses in the films is usually undesirable. Undesired crack formation or blistering can occur in the films. In the worst cases, the films can even become completely detached from the substrate. The observed total mechanical stress can consist of three components, which are the external stresses, thermal stresses, and intrinsic stresses. The external stresses arise from attack by external forces. Thermal stresses always arise if large differences exist in the thermal expansion coefficient between film material and substrate material. The last and most important part is the intrinsic stress in the film itself. Intrinsic stress is a structure- and microstructure sensitive property which is caused by the mode of film growth and microstructural interactions. Usually, the intrinsic stresses in the films are the dominant part of the total stress.

Fig. 5 illustrates the variation of mechanical stresses with changing the working point (changing the current setpoint). The stresses have been calculated in two perpendicular directions, and then a rounded arithmetic mean was plotted as a function of the DC current setpoint. The variation of the working point induces a large influence on the residual stress of the layers: The stress changes from tensile to compressive within the examined range of 14…20 A, which indicates that the residual stress can be tailored. No significant difference in the film stresses is noticed for the two samples deposited under same conditions, one with additional heating and the other without.

![Fig. 5: Working point dependent film stress](image)

3.3 Mechanical Properties
The mechanical properties of thin films produced by various deposition techniques are strongly dependent on structure, microstructure, and chemical composition, in particular, incorporated impurities. They can thus be influenced by the production technology used and the parameters chosen. Very few articles investigate the mechanical properties of AlN films. In this experimental work, we report the effect of changing the DC current setpoint on both the hardness and the modulus of Elasticity (Young’s modulus). A Nano-Indenter XP® from MTS was used to measure these mechanical properties (Poisson’s ratio of 0.3 assumed for the evaluation).

![Graph](a) & (b)

**Fig. 6: Working point dependent mechanical properties: (a) Hardness & (b) Young’s modulus**

Fig. 6 shows the mechanical properties of AlN films deposited at different working points. The error bars are obtained statistically from the measurements. The value for hardness tends to decrease with increasing the DC current setpoint from 14 to 16 A. However, it increases again up to 20 A current setpoint. Whereas, the value for the modulus of Elasticity decreases between 14 to 18 A setpoints, and then slightly increases from 18 to 20 A. In view of the limited number of points and of the size of the error bars, one can only state that the variation of the working point has only small effects on the mechanical properties of the layers. The hardness amounts to between 22 and 26 GPa. And Young’s modulus ranges from 309 to 336 GPa. The two samples, which were deposited at the same working point, with and without additional heating system, do not show significant differences in mechanical properties.

### 3.4 SEM Investigations

Scanning Electron Microscopy, DSM 962 (Zeiss), was used to acquire more information on the surface topography and film morphology of our AlN layers. Fig. 7 shows the fracture cross section of the two film samples deposited at 14 and 20 A setpoints, respectively.

Both layers have a pronounced columnar microstructure. The layer deposited at a more reactive working point (20 A setpoint) is characterized by a denser microstructure with a relatively smooth surface. This seems to be due to enhanced particle bombardment of the growing film. The columnar crystallites are interrupted several times in direction of layer growth. The grain size is relatively small and amounts to about 50 nm on the layer surface.
In contrast the layer deposited at a more metallic working point (14 A setpoint) has columnar crystallites which can be followed up through the whole layer. In addition, some indications of porosity can be recognized on the grain boundaries. The layer has a rougher surface with faceted crystallites and a higher lateral grain size up to 100 nm.

The observed differences in morphology of the layers can be correlated to the residual stress in the layers. i.e., compressive stresses develop with increasing the layer density.

4 CONCLUSIONS

A study for the reactive deposition of AlN thin films by bipolar dual magnetron sputtering, using a pure Al target in an ambient of Ar and N\(_2\) has been carried out. The respective process characteristic curves were recorded in both, a fixed flow mode and a current controlled mode. Moreover, 1.5 µm thick layers of AlN were deposited on (111) oriented Si substrates at different working points. The films were investigated to study the effect of varying the DC current setpoint of the controller on the resulting film properties, such as: the film thickness, residual stresses, and mechanical properties. In addition, scanning electron microscopy was used to examine the surface topography and film morphology at fracture surface. Also, an attempt has been made to correlate this structure to the obtained properties.

All films were stoichiometric and highly transparent. Mechanical properties determined on coated Si-Wafers showed that the films exhibit hardness up to 26 GPa and elastic modulus up to 336 GPa and these properties do not vary significantly with varying the current setpoint. In contrast, the residual stress can be tailored from +0.5 GPa tensile to −0.5 GPa compressive stress by appropriate choice of deposition conditions. SEM images revealed that a smoother layer with a denser structure results when working at higher current setpoint.
5 ACKNOWLEDGEMENT

One of the authors (M. Bedewy) would like to thank the Fraunhofer society for applied research (Fraunhofer-Gesellschaft zur Förderung der angewandten Forschung e.V., Germany) for the scholarship to complete this work at the Fraunhofer Institute for Electron Beam and Plasma Technology.

6 REFERENCES


