Thermal conductivity measurement of AlN films by fast photothermal method.

K. Ait Aissa¹, N. Semmar², D. De Sousa Meneses³, L. Le Brizoual¹, M. Gaillard², A. Petit², P-Y. Jouan¹, C. Boulmer-Leborgne² and M.A.Djouadi¹

¹ Université de Nantes, CNRS, Institut des Matériaux Jean Rouxel, UMR 6502, 2 rue de la Houssinière B.P. 32229 - 44322 Nantes cedex 3, France.
² GREMI, Groupe de Recherche sur l’Energie des Milieux Ionisés, UMR-7344, CNRS-Université d’Orléans, 14 rue d’Issoudun, BP 6744, 45067 Orléans Cedex2, France.
³ Conditions Extrêmes et Matériaux : Haute Température et Irradiation, UPR3079 CNRS, 1D Avenue de la Recherche Scientifique, 45071 Orléans Cedex.
E-mail: nadjib.semmar@univ-orleans.fr

Abstract. Aluminum nitride (AlN) films were deposited by reactive direct current Magnetron Sputtering (dcMS) on Si (100) substrates, with different thicknesses, in Ar–N₂ gas mixture. The films were characterized by X-ray diffraction (XRD), profilometry, scanning electron microscopy and UV-Visible Ellipsometry. The effect of the thickness on the thermal conductivity of AlN films was investigated using a fast IR pyrometry device. The XRD measurements show that AlN films are texturated along (002) direction. Moreover, X-ray rocking curve measurements indicate that the crystalline quality of the AlN is improved with the increase of film thickness. Optical analyses by IR spectroscopy and UV-Visible Ellipsometry demonstrate a high optical band gap of pure AlN films with semi-transparent behaviour in the IR range (1 to 7 µm). The effective thermal conductivity of the AlN films is strongly dependent on the film thickness. An effective thermal conductivities between (80 ± 0.5) and (175 ± 1.5) W.m⁻¹.K⁻¹ were measured for 260 and 8000 nm thick AlN film.

1. Introduction
Aluminum nitride is an important III-V material due to its unique thermal, mechanical and optical properties. It can be applied as active and passive layer in microelectronic devices, heat sinks in electronic packaging applications, component for acoustic wave devices and UV electroluminescent device [1]. It has been proposed as an alternative to alumina and Beryllia for thermal management application [2], because of its high thermal conductivity and small thermal expansion mismatch compared to that of silicon. The thermal conductivity of bulk AlN, at room temperature, along the c-axis can be as high as 320 W.m⁻¹.K⁻¹ [3], but it is usually considerably lower for polycrystalline AlN. This significant thermal conductivity reduction is caused by the presence of oxygen impurities [3-5]. Hence it has been shown that the thermal conductivity of AlN is inversely proportional to the oxygen content in the AlN lattice [3, 6]. In addition, the thermal conductivity of a thin film and bulk material can be substantially different because of the microstructure, including lattice impurities or defects [3], crystallite phase, grain boundaries, and porosity. Recently, the thermal conductivity value of AlN films deposited on silicon has been
raised up to 170 $W.m^{-1}.K^{-1}$, thanks to the improvement of crystalline quality and reduction of oxygen content in the films [7].

To date, many deposition techniques for AlN films have been reported including: Chemical Vapor Deposition (CVD) [1], and Molecular Beam Epitaxy (MBE) [8]. However, these methods are expensive and the required high temperatures to reach satisfying properties are often incompatible with microelectronic processes. Among other techniques, the Reactive Magnetron Sputtering process (RMS) is an attractive deposition technique because it presents advantages of being low temperature and low cost methods, and it allows fine tuning of the material characteristics [9-11].

This paper reports on the microstructure dependence of the thermal conductivity of AlN films. Those are deposited by dcMS, with a (002) single orientation along c-axis direction. The effect of the thickness on the thermal conductivity was investigated using the photothermal method in the nanosecond time domain.

2. Experimental procedure

2.1. Aluminum nitride deposition

Aluminum nitride films were deposited on Si (100) substrates, without intentional heating by dc reactive magnetron sputtering, using aluminum target (50 mm in diameter and 99.999 % purity), in an argon and nitrogen atmosphere, with a target-substrate distance of 30 mm. The sputtering pressure was fixed at 0.4 Pa. The residual pressure in reactor chamber was less than $6 \cdot 10^{-6}$ Pa. The total gas flux rate during deposition was maintained constant at 40 sccm while $N_2$ to Ar gas ratio was fixed at 35. An unbalanced magnetron sputtering system powered by a 150 W dc power supply was used. In order to check the effect of the thickness on the thermal conductivity, different samples were prepared by varying the film thickness from 260 up to 8000 nm.

The films were characterized by X-ray diffraction, using D5000 MOXTEK diffractometer with (Cu Ka) radiation ($\lambda = 0.1540598$ nm) in the $\theta - 2\theta$ Bragg Brentano and $\theta - \theta$ Rocking Curve configurations. The surface morphology and microstructural characterization were carried out by field emission gun scanning electron microscopy (ZEISS SUPRA 40). Refractive index (n) and extinction coefficient (k) were characterized with Ellipsometry measurements, performed with a phase modulated Ellipsometer Horiba Jobin Yvon UVISEL and infrared reflectivity measurements performed on a Bruker IFS 113 v spectrometer with an instrumental resolution of 4 cm$^{-1}$. The thicknesses of the films were estimated by using a DEKTAK VEECO 8 profiler.

2.2. Thermal measurement set-up

The thermal conductivity of AlN films was measured with a nanosecond pulsed photothermal method (PPT). The experimental set-up (Fig 1) was described in detail in Refs [12, 13]. This method consists of detection of the IR radiations emitted from the surface of a sample after its interaction with a pulsed-laser beam. The sample, being heated by a UV KrF laser pulse ($\lambda = 248$ nm, $\tau = 27$ ns), emits IR thermal waves that are focalized by two parallel off-axis parabolic mirrors into the active area (0.25 mm diameter) of an IR detector. The IR detector is cooled with liquid nitrogen and has a wide spectral range from 2 to 12 $\mu m$ (300 – 3000 $K$) with an integrated preamplifier of 100 $MHz$. The output signal of the IR detector is readed by a numerical oscilloscope (500 $MHz$ bandwidth) that can give simultaneously the time distribution of the KrF laser beam (UV) and the IR signal variations at the nanosecond time scale.
3. Results

3.1. Structural properties

![Figure 1. Schematic view of the fast IR pyrometry setup.](image)

**Figure 1.** Schematic view of the fast IR pyrometry setup.

**Figure 2.** X-Ray Diffraction patterns of AlN films deposited with different thicknesses.

**Figure 3.** Rocking curve FWHM (002) values of AlN layers as a function of the AlN film thickness.

The XRD diffraction patterns of AlN films deposited with different thicknesses are shown in Fig 2. The $\theta - 2\theta$ measurement exhibits a narrow peak with a full width at half maximum (FWHM) lower than 0.2°. The films exhibit a single orientation along c-axis direction, which is attributed to wurtzite-type structure of AlN. The effect of AlN film thickness on the FWHM values of the (002)-peak rocking curve is presented in Fig 3. It is shown that the FWHM value decrease with AlN film thickness. In fact, it appears clearly that the c-axis orientation (represented by the decrease of FWHM value) depend directly on the film thickness up to a value of approximately 6000 nm. Once this value is reached, there is no influence of the layer thickness on FWHM (002)-rocking curve values. Such results, which indicate an improvement of the crystalline quality with the increase of film thickness, have been obtained as a result of the use
of an unbalanced configuration of magnetron sputtering. This promotes high ion/neutral ratio and deposition energy in the range of 20 to 50 eV [14].

3.2. Optical properties

Several experiments were conducted on AlN thin films by Fourier Transform IR spectroscopy and UV-visible ellipsometry in order to cover the whole optical range, i.e. from 0.05 to 6 eV (402 to 48300 cm\(^{-1}\)). Fig 4a, reports the refractive index (n) and extinction coefficient (k) deduced from infrared reflectivity measurements. The optical response is typical of that of a dielectric material where the low wavenumber part, below 1000 cm\(^{-1}\), corresponds to the phonon response of AlN. The optical gap for which the material can be considered as nearly transparent (very low absorption coefficient) spans between 0.25 and 3 eV. Fig 4b contains the spectral evolution of the optical indices in visible and UV ranges obtained by ellipsometry, the spectral trends show that the electronic absorption starts to be significant only above 3 eV. In the near infrared and visible ranges, the refractive index of the AlN film is close to 2.1 a value corresponding to that of bulk AlN [15].

3.3. Thermal properties

For thermal measurement, the AlN films were coated by titanium (Ti) layer (Fig 5) which acts as an optical transducer to conversion the UV photon flux into IR radiation. In order to obtain absolute temperature evolution, it was necessary to calibrate the IR detector response. In this calibration process [16], the samples are heated to different levels of temperatures and for each temperature value; a detector output voltage is measured. The obtained calibration curve (Fig 6) is then used to transform the measured voltage (Fig 7a) to the temperature values (Fig 7b).
Fig 5. Show the cross section SEM image of 8000 nm thick AlN films coated with ∼200 nm thick Ti layer transducer. The AlN film shows a very compact and dense structure, without any void. Such structure can be associated to zone 2 of the Thornton materials diagram [17] even if these films were synthesized without intentional heating of the substrate (temperature < 250 °C). The film density promotes an abrupt interface with the post deposited Ti metallic coating as it is shown in the inset of Fig 5.

Typical result from the IR detector is plotted in Fig 7. in case of 200 nm Ti on 8000 nm thick AlN film. Using the calibration procedure, one should calculate the returned surface temperature in the nanosecond scale.

From those signals, a multilayer model [16], is used under Comsol Multiphysics to extract the conductivity values that allow the best fitting of the experiment signals. Because it is difficult to evaluate directly the thermal contact resistance due to AlN/Si interface, the computation
of the equivalent thermal resistance of the whole contributions (effective thermal conductivity) including the interface and the microstructure defects, gives a value close to $2 \cdot 10^{-9} \text{K.m}^2\text{W}^{-1}$.

Figure 8. (a) Effective thermal conductivity, - (b) Thermal resistance, as a function of the thickness AlN films.

Fig 8. shows the effective thermal conductivity and related thermal resistance as a function of thickness. Both thermal properties increase with the film thickness. According to the results of the XRD analysis it can be claimed that the increased thermal conductivity of AlN film from 80 to 175 $\text{W.m}^{-1}\text{K}^{-1}$ with increasing the film thickness from 260 to 8000 nm is primarily due to lower FWHM (002)- rocking curve which followed an increase in size grain size. In general, the size of a grain is proportional to the thickness of the thin film if other process conditions are unchanged [18] and the low thermal conductivity at 260 nm may be due to lattice mismatch at the interface of AlN/Si. The film interfacial regions near the substrate have generally a poor microstructure due to lattice mismatch [19]. This increasing of the thermal conductivity goes with increasing of the thermal resistance between all the AlN film and the Si substrate, with a value of $0.52 \cdot 10^{-8}$ to $4.8 \cdot 10^{-8} \text{K.m}^2\text{W}^{-1}$ for a thickness range from 260 to 8000 nm, this result agrees with that reported by C. Duquenne [7]. This thermal resistance is not only attributed to the interfacial amorphous layer between the AlN film and the substrate but also to defaults all along the film thickness.

To analyse the accuracy of such measurements the sensitivity of our IR method is evaluated at different thermal conductivity values: 50, 100 and 150 $\text{W.m}^{-1}\text{K}^{-1}$ as plotted in Fig 9. The maximum sensitivity is obtained in the time range of 5 to 10 ns, close to the maximum input laser beam. For example, considering a sensitivity of 0.4 $\text{W}^{-1}\text{m.K}^2$, which corresponds to a surface temperature of 600 K, one could evaluate from the calibration curve, the variation of the output voltage for 1 K deviation : $\delta U/\delta T = 4a_{4}T^3 \simeq 0.3 \text{mV.K}^{-1}$. From those values, the variation of output voltage versus thermal conductivity is roughly 0.12 $\text{mV.W}^{-1}\text{m.K}$. That means 1 $\text{mV}$ deviation is corresponding to less than 10 $\text{W.m}^{-1}\text{K}^{-1}$ thermal conductivity variations. In evidence higher is the temperature level; better is the the sensitivity to the thermal conductivity changes.

However, in this case, such estimation is conducted on a wide temperature range (300 to 650 K)
in the present work). First measurement done one 1900 \textit{nm} AlN films at different temperature levels (300, 350...550 K) have shown a very slight variation of the output voltage leading to quasi-constant values of the effective thermal conductivity (Fig 10).

![Figure 9. Sensitivity ($\delta T/\delta k$) versus time plotted for 50, 100 and 150 W.K$^{-1}$m$^{-1}$ AlN thin films.](image1)

![Figure 10. Normalized output signal from IR detector versus time at different W/AlN substrates temperatures from 300 to 550 K.](image2)

4. Conclusion
This study investigated the thermal conductivity of AlN films deposited by Reactive Magnetron Sputtering at low temperature, using unbalanced magnetron. It has been shown that the deposited AlN films are oriented along c-axis. The crystalline quality of the films was found to be improved with the film thickness, which in turn, affects the thermal conductivity of the material. The effective thermal conductivity value of AlN thin films increases from $60 \pm 5$ up to $175 \pm 15$ W.m$^{-1}$K$^{-1}$ for 260 and 8000 nm thick AlN films respectively, with a weak variation versus temperature in the range 300 to 550 K. These results highlight AlN films for thermal management applications.
References

[1] Radhakrishan G 1995 J. Appl. Phys. 10 1237.
[2] Melloch Micheal R, Adesida Ilesanmi, Isaacs-Smith T, Madangarli V and Sudarshan T.S 1997 J. Electron. Mater. 26 212.
[3] Slack G A, Tanzilli R A, Pohl R O and Vandersande J W 1987 J. Phys. Chem. Solids 48 647.
[4] Warti K, Ishizaki K and Fujikawa T 1992 J. Mater. Sci. 27 2630.
[5] Wartari K, Ishizaki K and Tsuchiya F 1993 J. Mater. Sci. 28 3714.
[6] Dinwiddie R B, Whittaker A J and Onn D J 1989 J. thermophys. 10 1084.
[7] Duquenne C, Besland M.P, Tessier P.Y, Gautron E, Scudeller Y and Averty D 2012 J. Phys. D 45 015301.
[8] Tanaka S, Kem R.S, Bentley J and Davis R.F 1996 J. Appl. Phys. 35 1641.
[9] Khanna A and Bhat D.G 2007 J. Vac. Sci. Technol. A 25 565.
[10] Drusedau T.P and Blasing J 2000 Thin Solid Films 378 31.
[11] Vergara L, Clement M, Iborra E and al 2004 Diam. Relat. Mater. 13 842.
[12] Amin-Chalhub E, Semmar N, Coudron L, Gautier G, Gailllard M, Petit A, Boulmer-Leborgne C and Millon E 2011 Journal of Physics D: Applied Physics D 44 355401.
[13] Martan J, Semmar N, Boulmer-Leborgne C, Plantin P and Le Menn E 2006 Nano. Microscale Thermophys. Eng. 10 12.
[14] Duquenne C, Popesco B, Tessier P.Y, Besland M.P, Scudeller Y, Brylinski C, Delage S and Djouadi M.A 2007 Plasma Process. Polym. 4 S1 S5.
[15] Legrand P.B, Wautelet M, Dugnoille B, Dauchot J.P and Hecq M 1994 Thin solid Films 248 220.
[16] Martan J, Semmar N, Leborgne C, Le Menn E and Mathias J 2005 Applied Surface Science 247 63.
[17] Thornton J.A 1977 Annual review of materials science 7 260.
[18] Mayadas A.F and Shatzkers M 1970 Phys. Rev. B 1 1382.
[19] Abdallah B, Duquenne C, Besland M P, Gautron E, Jouan P Y, Tessier P Y, Brault J, Cordier Y and Djouadi M A 2008 Eur. Phys. J. Appl. Phys. 43 309.