Reactive pulse magnetron sputtering for deposition of piezoelectric AlN layers

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Reactive pulse magnetron sputtering of Al targets in a gas mixture of Argon and Nitrogen allows the deposition of AlN layers at high deposition rates of up to 200 nm/min. In the reported experiments films were deposited AlN onto unheated substrates with a thickness of typically 10 µm. Deposited films have been characterized for a variety of layer properties using e.g. XRD, SEM, profilometry, weighting and piezoelectric measurements regarding crystalline structure and orientation, surface morphology, density, film stress and piezoelectric coefficient d_{33}. The characterized AlN films can be classified into 2 groups. The first group shows a nearly pure 001 orientation of the crystalline structure, an undisturbed surface morphology, a high density and a very high piezoelectric coefficient d_{33} of up to 7.2 pm/V on silicon substrate. The second group exhibits a dominating but not pure 001 orientation, disturbances in the surface morphology, a slightly lower density and a piezoelectric coefficient close to zero. The range of the process parameters pulse mode, pressure, sputtering power and reactive working point to achieve the layers of the first group is very narrow. Surprisingly films with high piezoelectric constant can be obtained both by strong and moderate particle bombardment during deposition using adapted parameter sets. The suitability for the intended application in high frequency ultrasonic phased array sensors systems is investigated using pulse echo measurements.

Keywords: Reactive pulse magnetron sputtering, AlN, Piezoelectric layers, Ultrasound

I. INTRODUCTION

New 3-dimensional integrated electronic devices, next generations of textured materials like carbon fiber reinforces polymers (CFRP) and complex multilayered devices like solar cells require new approaches for non destructive evaluation techniques. One area of research and development are high frequency ultrasonic phased array sensors systems that combine the performance of phased array methods with the resolution of a scanning acoustic microscope. The very high frequencies in phased array sensors require a very thin transducer thickness and cannot be realized with traditional piezoelectric materials like PZT ceramics, piezoelectric 1-3 composites or polymers like polyvinylidenfluoride (PVDF). A promising alternative piezoelectric material is aluminum nitride (AlN).

Aluminum nitride is a piezoelectric but not ferroelectric material with a Wurtzite crystal structure. Compared to the widely used ferroelectric materials like PZT, AlN cannot be electrically polarized. Therefore piezoelectric activity can only be observed in single crystals or in a polycrystalline structure with a strong crystal orientation. To achieve a thickness vibration of the sensor, a crystalline orientation in (001) direction is necessary (c-axis of the AlN crystalline structure being oriented perpendicular to the substrate surface).

In this paper, piezoelectric AlN films with a thickness of up to 10 µm were investigated. The reactive pulse magnetron sputtering process was optimized for high rate deposition of AlN thin films with strong piezoelectric properties.

II. EXPERIMENTAL

Coatings were carried out in cluster type sputter equipment using the Double Ring Magnetron DRM 400 developed at Fraunhofer FEP. This type of magnetron combines two concentric discharges allowing uniform coating of substrates with a diameter up to 200 mm [1]. Figure 1 shows the schematic of the deposition set up. Pulse powering at 50 kHz in unipolar or bipolar pulse mode was applied using the pulse unit UBS-C2 of Fraunhofer FEP and standard DC power supplies. Pure metallic aluminum targets were sputtered in a mixture of argon and oxygen as reactive gas. A closed loop control of the reactive gas inlet allowed stabilizing the process in the so-called transition mode, where stoichiometric films are deposited at high deposition rates.

Using the DRM 400, the pulse mode of the pulse magnetron sputtering process can be changed between unipolar and bipolar. In the unipolar pulse mode, a pulsed dc is applied between each of the two targets and the separate hidden anode. In the bipolar pulse mode, a voltage with alternating polarity is applied between the two targets. For the two pulse modes, the plasma properties are completely different (Table I). Thus, variation of the pulse mode allows new degree of freedom to optimize deposition process. On one hand, the unipolar pulse mode shows low thermal substrate load and it allows e.g. coating of very temperature sensitive substrates. On the other hand, in the bipolar pulse

 ISBN: 978-1-61208-145-8

mode a very intense energetic ion bombardment occurs that allows deposition of very dense films.

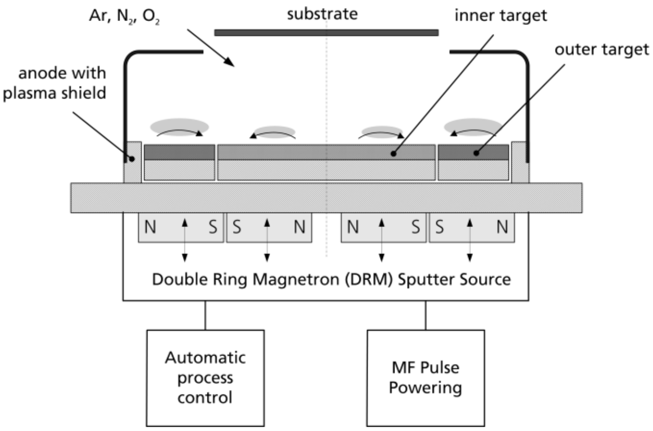

Figure 1. Deposition setup for sputter deposition

TABLE I. RESULTS OF LANGMUIR PROBE AND TEMPERATURE MEASUREMENTS, SiO_2 SPUTTERING AT 7.5KW

Pulse mode		unipolar	bipolar
Plasma density	**[1/cm^3]**	$1.8 \cdot 10^{10}$	$11.0 \cdot 10^{10}$
Electron temperature	**[eV]**	10	6
Thermal substrate load	**[W/cm^2]**	0.15	0.75

From literature it is known, that the sputter parameters strongly influence the piezoelectric behavior [2]. Thus, for each of the two pulse modes the power and voltage applied to the sputter target as well the pressure was varied to find optimal process parameters for the deposition of AlN layers with highest piezoelectric properties. In Table II, deposition parameters are summarized.

TABLE II. RANGE OF ALN DEPOSITION PARAMETERS

Sample		AlN-UP	AlN-BP
Pulse mode		**unipolar**	**bipolar**
Preheating		**room temperature**	**room temperature**
Power	**[kW]**	**6…13**	**6…13**
Pressure	**[Pa]**	**0.15…2**	**0.15…2**
Deposition rate	**[nm/min]**	**100...200**	**80...160**

For the evaluation of piezoelectric properties, a simple ultrasound transducer test layout was used. An electrode structure with 10 mm diameter was deposited on an isolated silicon wafer. An AlN film in a circle structure with a diameter of 13 mm was deposited, followed by a second aluminum electrode to fabricate the sensor and the interconnection pad on the top side. The structures were realized by using laser cutted aluminum oxide ceramic masks with a high stiffness to prevent bending when being reused. The area of the deposited sensor layer needs to be larger than the electrodes themselves, because there occur edge effects during the deposition process, which influence the piezoelectric behavior of the sensor.

III. STRUCTURAL, ELECTRICAL AND MECHANICAL PROPERTIES

The characterized AlN films can be classified into 2 groups. The first group shows a nearly pure 001 orientation of the crystalline structure, an undisturbed surface morphology, a high density and a very high piezoelectric coefficient d_{33} of up to 8 pm/V. The second group exhibits a dominating but not pure 001 orientation, disturbances in the surface morphology, a slightly lower density and a piezoelectric coefficient close to zero. The range of the process parameters pulse mode, pressure, sputtering power and reactive working point to achieve the layers of the first group is very narrow. Surprisingly films belonging to the first group with high piezoelectric constant can be obtained both by moderate and strong particle bombardment during deposition in unipolar and bipolar pulse mode respectively.

Figure 2 shows the XRD diagrams of films with nearly pure 001 orientations (002 and 004 peaks) deposited in unipolar and bipolar mode. Figure 3 shows the results of SEM investigations on these samples. The fracture in the SEM micrograph exhibits in both cases a dense microstructure. Surface morphology is rather coarse in bipolar compared to unipolar pulse mode. In Table III, most important layer properties for the two layers with highest piezoelectric properties are summarized. Films deposited in bipolar mode exhibit slightly higher values of piezoelectric coefficient, density, resistivity and breakdown field strength, but show significantly stronger compressive stress.

TABLE III. DEPOSITION PARAMETERS AND LAYER PROPERTIES FOR PIEZOELECTRIC ALN LAYERS

Sample		AlN-UP	AlN-BP
Pulse mode		Unipolar	Bipolar
Crystalline orientation (fraction)		002 (99.9%)	002 (99.9%)
Density	**[g/cm3]**	3.16	3.20
Break down field strength	**[MV/cm]**	2.3	3.1
Resistivity	**[Ωcm]**	$5.3 \cdot 10^{12}$	$1.2 \cdot 10^{13}$
piezoelectric charge constant d33	**[pm/V]**	6.5	7.2
Mechanical stress	**[GPa]**	-1	-2

 ISBN: 978-1-61208-145-8

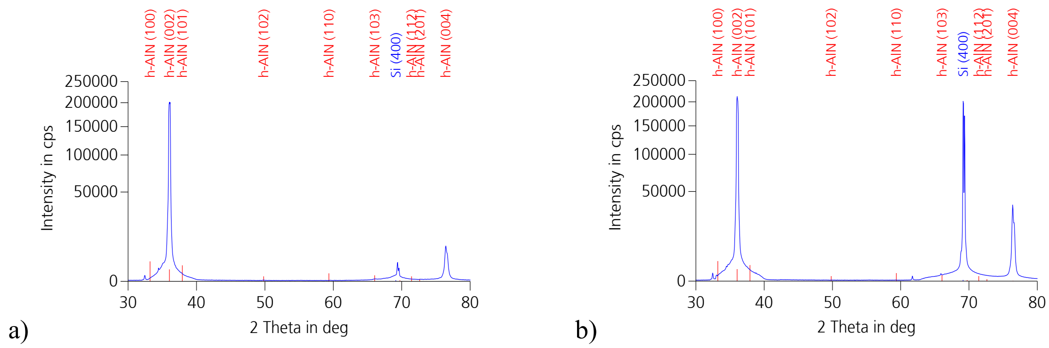

Figure 2. XRD diagrams (a: unipolar pulse mode, b: bipolar pulse mode)

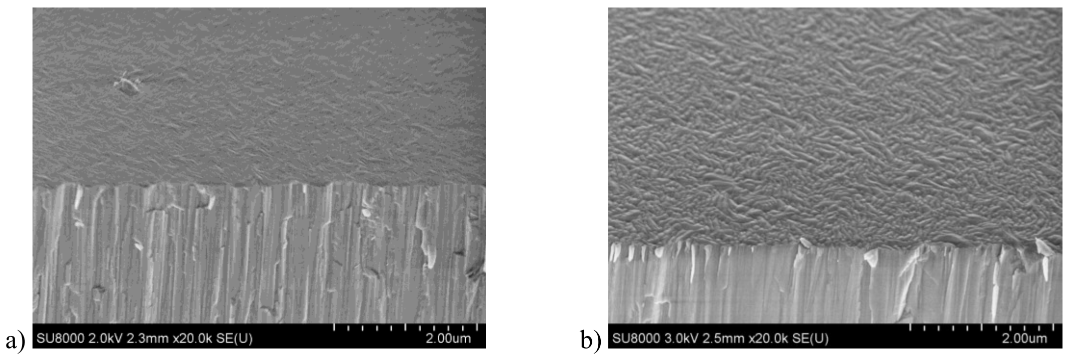

Figure 3. SEM-micrographs of AlN films, thickness 10µm (a: unipolar pulse mode, b: bipolar pulse mode)

IV. CHARACTERIZATION OF PIEZOELECTRIC PROPERTIES

The AlN thin films were used already for SAW filters, microwave filter or resonator and ultrasound transducers [3-5]. One possible new application is as sensor thin films in special phased array ultrasound transducers. For this application, the transformation of electrical energy in acoustical energy and vice versa has to be verified and quantified. Acoustical measurement in pulse echo mode were carried for each sensor to characterize the vibration behavior and the maximum signal voltage. Afterwards the electrical properties and piezoelectric charge constants were measured with a Berlincourt-Meter.

A. Pulse Echo Mode

In the pulse echo measurements the AlN sensor serves as an acoustic transmitter and receiver. The pulser and receiver DPR 500 (JSR Ultrasonics) was used to excite an acoustic sound wave. The ultrasound wave propagates through the silicon substrate, is reflected at the interface silicon-air and travels back to the AlN layer. The aluminum metal electrode with 150 nm thickness has only a very small influence and can be neglected. The mechanical vibration pulse gets transformed to an electrical signal, which can be measured.

The illustration in Figure 4 shows the schematic setup of the pulse echo measurements. All AlN sensors were connected to the pulse generator and were excited with a needle pulse at high amplitude (-143 V) and very short pulse time (~1.4 ns). The receiver was set to a gain of 36 dB and a high frequency pass filter between 30 MHz and 500 MHz was used.

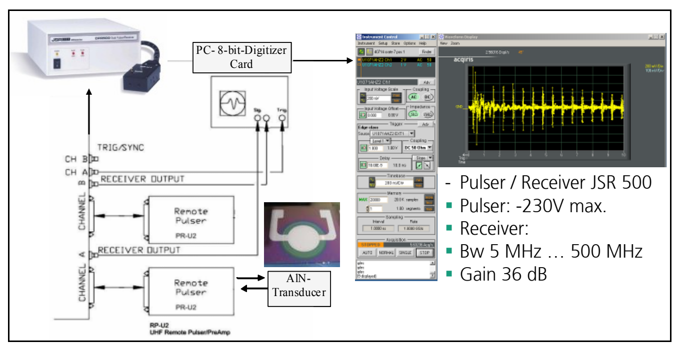

Figure 4. Measurement setup with Pulser/Receiver and PC Digitizer Card.

In the first tests the receiver output was connected to an oscilloscope, but during the evaluation of all AlN sensors the receiver signals where transmitted to a PC Digitizer Card (Aquiris U1071 A, Agilent Technologies). This hardware has an real-time sampling rate up to 2 GS/s and the amplifier response are optimized to ensure that high-frequency measurements can be made with a bandwidth (-3 dB) of maximum 1 GHz. The sending pulse after excitation could be observed, followed by multiple reflections from the silicon back wall. The distance between the multiple echoes equals the time the ultrasound longitudinal wave needs to pass through the silicon and return back. Because of the small damping behavior of silicon, the echo pulse signal is relatively long. The evaluated sensors were deposited on silicon substrates with the same thickness and were excited with the same electrical pulse. Therefore the amplitudes of the received signals could be used to compare the sending-receiving efficiency of the different AlN transmitters indirectly.

In the right handed diagram of Figure 4, a typical time response with multiple back wall echoes is shown. To avoid an influence of the sending signal, the first back wall echo was not evaluated, but the fourth. In Figure 5, the fourth back wall echoes are shown for the optimized layers deposited in the unipolar and bipolar pulse mode as well as for a not optimized layer. Strong differences in the transmitting and receiving properties of AlN based sensor could be verified as result from the different sputtering and deposition parameters.

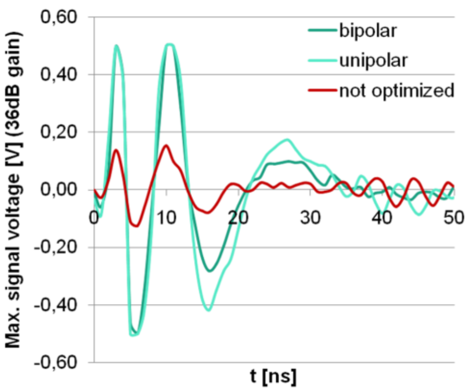

Figure 5. Examples of signal amplitudes of one back wall echo depending on the deposition parameter set.

B. *Berlincourt-Meter*

Additionally the piezoelectric charge constant (d_{33}) was measured with a Berlincourt piezometer PM 300 (PiezoTest). The samples were clamped and loaded with an alternating force. The generated electric charge was compared to the value of a reference sample to obtain the piezoelectric charge constant. The measurements were carried out by applying an alternating force of 0.25 N and a frequency of 110 Hz. Quasi-static measurements of the dissipation factor tan δ and the electric capacity of the AlN sensor layers was performed at a frequency of 1 kHz.

Measured Samples piezoelectric charge constant $d_{33}$for AlN thin films was between below 1 pC/N and 7.2 pC/N in maximum. The reasons for these differences is the different microstructure and c-axis orientation of the AlN thin films. Figure 6 shows, that the d_{33} value can be correlated to the maximum signal voltage obtained in the pulse echo measurements. Therefore the Piezometer can be used for a very fast estimation of the thin film quality for AlN sensors with the same substrate material and film thickness.

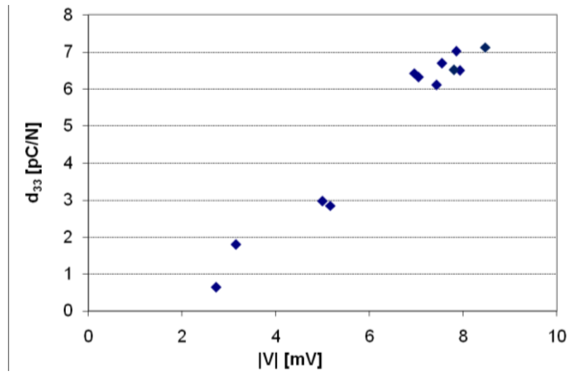

Figure 6. Correlation of maximum pulse amplitude and piezoelectric charge constants.

V. CONCLUSIONS

In this paper, the piezoelectric behavior of ultrasound sensors based on the AlN thin films was investigated. The process parameters like pulse mode during sputtering, power and process pressure strongly influenced the crystalline growth and the orientation of the thin films. As result, sensor samples with excellent c-axis orientation were obtained, that were investigated regarding their structural, electrical and mechanical properties. The electro acoustic measurements and the evaluation of the piezoelectric charge constants have shown good piezoelectric activity. But, the piezoelectric activity is depending strongly on the deposition parameters. For AlN thin films on silicon substrates, d_{33} was about 7.2 pC/N in maximum for films with nearly perfect c-axis orientation and it was below 1.0 pC/N for films with weak c-axis orientation. Also in pulse echo measurements, the AlN sensors with good c-axis orientation showed a much higher echo amplitude.

ACKNOWLEDGMENT

This work has been partially supported by a Grant-in-Aid for Technology Funding by the European Regional Development Fund (ERDF) 2007-2013 and the State of Saxony.

REFERENCES

[1] P. Frach, Chr. Gottfried, H. Bartzsch, K. Goedicke, “The double ring process module-a tool for stationary deposition of metals, insulators and reactive sputtered compounds”, Surf. Coat. Technol. 90, 1997, 75-81.

[2] E. Iborra, A. Sanz-Hervás, M. Clement, L. Vergara, J. Olivares, and J. Sangrador, “Assessment of the piezoelectric response of sputtered AlN films by X-ray diffraction”, Proc. of the IEEE Ultrasonics Symposium , 2005, pp. 1808.

[3] M.-A. Dubois, P. Muralt, “Properties of aluminum nitride thin films for piezoelectric transducers and microwave filter applications”, Appl. Phys. Lett., Vol. 74, No. 2, 17 May 1999, pp.3032.

[4] L. Valbin, L. Sevely, “Piezoelectric aluminum nitride thin film for ultrasonic transducers”, Proc. of SPIE , Vol. 4559, 2001.

[5] P.M. Martin, M.S. Good, J.W. Johnston, G.J. Posakony, L.J. Bond, S.L. Crawford, ”Piezoelectric films for 100-MHz ultrasonic transducers”, Thin Solid Films 379, 2000, p 253-258.

 ISBN: 978-1-61208-145-8