Universidad Politécnica de Madrid

**Escuela Técnica Superior de Ingenieros de Caminos, Canales y
Puertos**

ISOM

Surface acoustic wave (SAW) devices on ScAlN thin films

Doctoral thesis

Miguel Sinusia Lozano

Director:

Gonzalo Fuentes Iriarte

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A mis padres y hermanas, Beatriz, familiares y amigos

ABSTRACT

The massive volume of data transfer rate in the novel 5G technology implies multiple requirements the electroacoustic devices should meet. For tackling these technological constraints, surface acoustic wave (SAW) devices have been proposed because of their outstanding performances such as power handling capabilities, temperature stability or achievable resonance frequencies. However, long-term studied piezoelectric thin films such as aluminum nitride (AlN) or thin oxide (ZnO) lack the necessary piezoelectric constants and thus the achievable electromechanical coupling coefficients to fabricate SAW devices meeting the 5G bandwidth requirements. Introducing scandium (Sc) atoms into the wurtzite AlN phase has reported the piezoelectric response enlargement of the scandium aluminum nitride (ScAlN) compound as compared to the pristine AlN, providing the foundation to fabricate 5G ScAlN based SAW devices. The possibility of employing the mature AlN synthesis processes expands even further the industrial and research attraction of the ScAlN compound.

The work carried out during these 3 years aims to study the feasibility of employing ScAlN thin films in 5G electroacoustic applications. The synthesis processes to obtain a reliable procedure promoting highly c-axis oriented ScAlN thin films together with the device design and fabrication routes have been studied in detail.

In order to cope with the thermal budget limitations of nowadays high throughput CMOS compatible manufacture schemes, reactive sputtering without intentional substrate heating has been the method used to deposit the ScAlN compound.

Since the piezoelectric response of the ScAlN compound is highly dependent on the Sc content, two alloy targets with different compositions, $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$, have been used during this thesis.

Several key parameters in the reactive sputtering process such as the discharge power or the process pressure have been studied to obtain highly c-axis oriented thin films on various substrates such as polycrystalline diamond. The unique presence of (0002) and (0004) reflections in X-ray diffraction θ -2 θ scans together with ω -scan full width at half maximum (FWHM) values of the (0002) reflection below 2 degrees assess the quality of our highly textured thin films. Compositional analyses reported 26% and 43% Sc content in layers synthesized using $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy targets, respectively. Measurements of the piezoelectric properties of our material confirm an increase of the piezoelectric d_{33} constant with the Sc content, as the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film duplicates (-24 pC/N) the d_{33} constant value obtained for the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film.

The SAW propagation characteristics of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ compounds on different substrates have been studied. In some cases, these characteristics agree with reported theoretical studies. However, the various improvements carried out during this thesis both in device design as well as from a materials perspective reflect on the electrical response of our SAW devices. As an example, the aforementioned increase of the d_{33} constant in the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film translates into an enhanced effective electromechanical coupling coefficient (K_{eff}^2). Rayleigh and Sezawa mode K_{eff}^2 values are increased by 650 % and 150% respectively in devices comprising $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films as compared to K_{eff}^2 values obtained for the same modes but being generated in $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ layers.

Finally, SAW filters have been fabricated on $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /diamond heterostructures with Sezawa mode bandwidths above 100 MHz and center frequencies close to 5 GHz. Our results confirm that ScAlN based devices are promising candidates to be implemented in 5G technology standards.

RESUMEN

La elevada cantidad de transferencia de datos de la nueva tecnología 5G implica que los dispositivos electroacústicos deban satisfacer ciertas especificaciones. Los dispositivos de ondas acústicas de superficie (SAW por sus siglas en inglés) son candidatos a ser utilizados en dispositivos para 5G debido a la capacidad de trabajar a elevadas potencias, su elevada estabilidad térmica y las frecuencias en las que son capaces de trabajar. Sin embargo, las capas piezoeléctricas delgadas que han sido estudiadas durante los últimos años como el nitruro de aluminio (AlN) o el óxido de cinc (ZnO) no poseen la respuesta piezoeléctrica necesaria para alcanzar el factor de acople electromecánico (K^2) para satisfacer el ancho de banda en la tecnología 5G. La inclusión de átomos de escandio (Sc) en la fase wurtzita del AlN incrementa la respuesta piezoeléctrica del compuesto resultante (ScAlN), situándolo como una gran opción para aplicaciones electroacústicas. Además, el hecho de poder utilizar las tecnologías desarrolladas para la síntesis de AlN expande todavía más el atractivo industrial y tecnológico del compuesto ScAlN.

El objetivo del trabajo llevado a cabo durante estos 3 años es el estudio de las propiedades del compuesto ScAlN para fabricar dispositivos para 5G. Por tanto, los procesos de síntesis para obtener capas delgadas altamente orientadas en el eje *c*, así como la fabricación y diseño de los dispositivos deben ser tenidos en cuenta.

En cuanto a la síntesis del compuesto, el proceso de pulverización catódica (sputtering) reactiva sin calentar intencionadamente el sustrato ha sido estudiado. Debido a la dependencia de la respuesta piezoeléctrica del compuesto ScAlN con el contenido de Sc, dos blancos con composiciones diferentes han sido empleados: $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$. La influencia que tienen los parámetros de la pulverización catódica reactiva, especialmente la presión y la potencia, han sido estudiadas para promover la síntesis de capas delgadas de ScAlN altamente orientadas en el eje *c* sobre diferentes sustratos como por ejemplo, diamante policristalino. La presencia en los análisis θ -2 θ de difracción de rayos X (XRD) de las reflexiones (0002) y (0004) del compuesto junto con los bajos valores del ancho a media altura del análisis ω ($\sim 2^\circ$) sobre la reflexión (0002) confirman la elevada textura de las capas sintetizadas. Los análisis de composición reflejan que los compuestos sintetizados con los blancos $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$ tienen una composición de $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ y $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$, respectivamente. Las medidas de la respuesta piezoeléctrica confirman el aumento que la constante piezoeléctrica d_{33} con el contenido de escandio, ya que la constante d_{33} en la capa delgada $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ (-24 pC/N) duplica a la medida en la capa $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$.

Las características de propagación de la onda acústica de superficie de ambos compuestos han sido estudiadas mediante dispositivos fabricados sobre diferentes sustratos. Estas características confirman los estudios teóricos presentes en la literatura, así como las diferentes mejoras llevadas a cabo durante la tesis sobre los diseños y materiales de los dispositivos se ven reflejadas en la respuesta eléctrica de los mismos. El comentado aumento de la constante d_{33} en la capa $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ se observa también en el factor de acople electromecánico (K_{eff}^2) de los dispositivos. Estos valores para los modos Rayleigh y Sezawa aumentan en un 650 % y en un 150% respectivamente, los valores K_{eff}^2 obtenidos en dispositivos fabricados con la capa $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$.

Finalmente, filtros de ondas acústicas de superficie han sido fabricados sobre una estructura con una capa $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ sintetizada sobre un sustrato de diamante. El ancho de banda del modo Sezawa en estos filtros es cercano a 200 MHz con frecuencias de resonancia cercanas a 5 GHz, lo que confirma que estos dispositivos pueden ser empleados en la tecnología 5G.

Table of contents

List of figures.....	xv
List of tables.....	xix
Agradecimientos.....	xxi
Publications.....	xxiii
I Introduction.....	1
1 Piezoelectricity.....	2
2 Surface acoustic waves (SAW) devices.....	3
3 Scandium aluminum nitride (ScAlN) compound.....	7
4 Aim of the thesis.....	8
5 Structure of the thesis.....	9
II Experimental.....	11
1 Reactive sputtering.....	11
1.1 Background.....	11
1.2 Reactive sputtering.....	12
1.3 Synthesis of c-axis ScAlN thin films by reactive magnetron sputtering.....	12
1.4 Influence of the sputtering parameters.....	13
2 Synthesis and device fabrication.....	15
2.1 Home-built reactive sputtering system.....	15
2.2 Synthesis conditions.....	16
2.3 Substrate cleaning.....	18
2.4 Sputtering system conditioning.....	18
2.5 IDT fabrication.....	19
3 Structural characterization.....	19
3.1 X-ray diffraction (XRD).....	19
4 Surface characterization.....	21
4.1 Scanning electron microscopy (SEM).....	21
4.2 Atomic force microscopy (AFM).....	21
5 Compositional characterization.....	22
5.1 Energy dispersive X-ray spectroscopy (XEDS).....	22
5.2 Secondary-ion mass spectrometry (SIMS).....	22
5.3 Rutherford backscattering spectrometry (RBS).....	23
5.4 Time of flight elastic recoil detection analysis (ToF-ERDA).....	24

6 Piezoelectric characterization.....	24
6.1 Piezoelectric force microscopy (PFM).....	24
6.2 Berlincourt piezometers.....	25
7 Electrical characterization.....	25
1.1 Electrical properties of SAW devices.....	26
1.2 Modified Butterworth van Dyke (mBVD) model.....	27
III ScAlN thin films: Results and discussion.....	29
1 Compositional analyses: on the Sc shortage between the thin film and alloy target compositions	29
2 Synthesis of highly c-axis oriented $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.....	33
2.1 On the influence of the gas admixture ratio in the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.....	36
2.2 Evaluating the rocking curve FWHM values: a comparative analysis.....	37
3 Piezoelectric analyses.....	40
3.1 $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films.....	40
3.2 $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.....	41
4 Summary.....	42
IV ScAlN based SAW devices.....	45
1 One port SAW resonators on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ based heterostructures.....	45
1.1 Influence of the ScAlN thin film thickness.....	46
1.2 Temperature coefficient of frequency (TCF).....	49
2 SAW devices on $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ heterostructures.....	51
2.1 One port SAW resonators.....	51
2.2 π type SAW filters.....	59
3 Summary.....	62
V Conclusion.....	65
VI Future work.....	67
VII References.....	71

List of figures

Figure I.1 Scheme of a single-electrode interdigital transducer (IDT) with reflectors at both ends.....	5
Figure I.2 One-port resonator configurations a) IDT design without weighting; b) IDT design with a double electrode transducer weighting; c) IDT design with apodized weighting; d) IDT design with weight weighting.....	6
Figure I.3 SAW based filter designs a) Double mode SAW (DMS) filter; b) Z-path SAW tag; Scheme of a c) ladder d) bridge electrical connection.....	7
Figure II.1 Different sputtering target configurations. Left: Two targets for a co-sputtering configuration. Right: Aluminum single target with scandium inlets in the race-track.....	13
Figure II.2 Home-built sputtering system employed for the synthesis of ScAlN thin films.....	16
Figure II.3 Lift-off process employed for the fabrication a SAW device IDT on a ScAlN/polycrystalline diamond heterostructure.....	19
Figure II.4 Left: Scheme of the Bragg Brentano geometry for XRD analysis. Right: Visualization of the Bragg equation.....	20
Figure II.5 Scheme of several energies produced by the interaction of the focused electron beam. Secondary electrons are recorded by the SEM equipment, whereas the energy dispersive X-ray spectroscopy (XEDS) technique relies on detecting the characteristic X-rays.....	21
Figure II.6 Scheme of the AFM equipment.....	22
Figure II.7 Left: Scheme of the SIMS equipment. Right: Schematic representation of the SIMS process.....	23
Figure II.8 Scheme of the RBS and ERDA analysis angles.....	24
Figure II.9 Scheme of a Berlincourt piezometer.....	25
Figure II.10 mBVD model implemented in the Keysight advanced design system (ADS). It simulates three different resonance modes with three discrete motional branches.....	27
Figure III.1 Composition of synthesized ScAlN thin films as measured from different analysis techniques. Left: $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films synthesized with the $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target. Right: $\text{Sc}_{0.43}\text{Al}_{0.57}$ thin films synthesized with the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target.....	30
Figure III.2 (Adapted from <i>Figure 3 of PAPER 1</i>) XEDS analysis of an as-synthesized $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films on B-doped Si (001) substrate.....	31
Figure III.3 SIMS analysis of an as-synthesized $\text{Sc}_{0.41}\text{Al}_{0.59}\text{N}$ ScAlN thin film on a polycrystalline diamond substrate.....	31
Figure III.4 RBS analysis of a 2000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a polycrystalline diamond substrate.....	32

Figure III.5 ToF-ERDA analysis of a 600 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a polycrystalline diamond substrate.....	33
Figure III.6 θ -2 θ analysis of a 100 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film on a polycrystalline diamond substrate.....	34
Figure III.7 Influence of the process pressure and the discharge power on the FWHM of ScAlN thin films synthesized on B-doped Si (001) substrates using two different alloy targets.....	35
Figure III.8 2 SEM cross section of a 2000 nm thick $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film synthesized on a polycrystalline diamond substrate. $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film synthesized with a process pressure of 0.40 Pa and a discharge power of 700 W.....	36
Figure III.9 Deposition rate as a function of the gas admixture ratio. These syntheses were carried out at a discharge power of 500 W and a process pressure of 0.4 Pa using the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target	36
Figure III.10 Influence of the gas admixture ratio in the rocking curve FWHM value of 1000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrates.....	37
Figure III.11 Comparison between the rocking curve FWHM values and substrate temperature during the synthesis. Substrates: Si (001): This thesis, PAPER 1, PAPER 2, [8], [42], [43], [46], [69], [70], [94], [95], [96], [97]; Al_2O_3 (0001): [97], [98], [99]; Pt (111): [100], [101]; Mo (110): [44] , [71], [100], [102]; TiN (111): [50], [90]; Ti (0002): [50]; AlCu (111): [50]; Polycrystalline diamond: This thesis, PAPER 3, PAPER 4, [103]. Broken parallel lines indicate overlapping of the adjacent points, which have been horizontally displaced.....	39
Figure III.12 (Above: Adapted from <i>Figure 6</i> of PAPER 2) 1x1 μm PFM analyses on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ (above) and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ (below) thin films synthesized on As-doped Si (001) substrates.....	40
Figure III.13 Influence of the gas admixture ratio in the piezoelectric constant d_{33} of 1000 nm and 300 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrates.....	41
Figure III.14 Maximum measured d_{33} constant of 1000 nm thick $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrate.....	42
Figure IV.1 Scheme of the SAW resonator fabricated using a Pt IDT on a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructure. IDT wavelength $\lambda=2800$ nm.....	46
Figure IV.2 (Adapted from <i>Figure 4</i> of PAPER 4) Reflection coefficient of SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Superimposed: mBVD simulation result. Inset: Impedance Smith chart. d stands for the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film thickness.....	47
Figure IV.3 (Adapted from <i>Figure 5</i> of PAPER 4) Effective SAW velocity of the Rayleigh and Sezawa modes generated in SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Lines are a guide for the reader.....	48
Figure IV.4 (Adapted from <i>Figure 6</i> of PAPER 4) Effective electromechanical coupling coefficient of the Rayleigh and Sezawa modes generated in SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Lines are a guide for the reader.....	48
Figure IV.5 (Adapted from PAPER 3) Frequency shift of the Sezawa mode reflection coefficient due to the temperature variation of SAW resonators fabricated on 2000 nm	

Sc _{0.26} Al _{0.74} N/polycrystalline diamond heterostructures. Above: First; Below: Second temperature iteration measurements.....	49
Figure IV.6 (Adapted from <i>Figure 5</i> of PAPER 3 ¹⁹) Temperature variation of the Sezawa mode effective acoustic velocities of a SAW resonator fabricated on 2000 nm Sc _{0.26} Al _{0.74} N/polycrystalline diamond heterostructures. Lines are a guide for the reader.....	50
Figure IV.7 (Adapted from <i>Figure 9</i> of PAPER 3 ¹⁹) Sezawa mode temperature coefficient of frequency of SAW resonators fabricated on 2000 nm Sc _{0.26} Al _{0.74} N/polycrystalline diamond heterostructures. Lines are a guide for the reader.....	51
Figure IV.8 Schematic illustration of the SAW devices fabricated using three different Sc _{0.43} Al _{0.57} N based heterostructures. IDT wavelength $\lambda=2800$ nm.....	52
Figure IV.9 Reflection coefficient of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/SiO ₂ heterostructure.....	53
Figure IV.10 Admittance characteristics of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/SiO ₂ heterostructure.....	54
Figure IV.11 Reflection coefficient of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/polycrystalline diamond heterostructure.....	55
Figure IV.12 Admittance characteristics of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/polycrystalline diamond heterostructure.....	56
Figure IV.13 Reflection coefficient of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/single crystal diamond heterostructure.....	57
Figure IV.14 Admittance characteristics of a SAW resonator fabricated on the Sc _{0.43} Al _{0.57} N/single crystal diamond heterostructure.....	58
Figure IV.15 Effective electromechanical coupling coefficient of the Rayleigh and Sezawa modes obtained in the polycrystalline diamond heterostructures with 2000 nm thick Sc _{0.26} Al _{0.74} N and Sc _{0.43} Al _{0.57} N thin films.....	59
Figure IV.16 Schematic of the π type SAW filters.....	60
Figure IV.17 Schematic illustration of the SAW filters fabricated using Sc _{0.43} Al _{0.57} N based heterostructures. IDT wavelength $\lambda=1200$ nm.....	60
Figure IV.18 Transmission coefficient of a SAW filter fabricated on a Sc _{0.43} Al _{0.57} N/polycrystalline diamond heterostructure.....	61
Figure IV.19 Transmission coefficient of a SAW filter fabricated on a Sc _{0.43} Al _{0.57} N thin film/single crystal diamond heterostructure.....	62
Figure VI.1 Transmission coefficient of a SAW filter fabricated with IDT of 65 nm Au and 5 nm of Cr as adhesion layer on a 200 nm Sc _{0.43} Al _{0.57} N thin film/polycrystalline diamond heterostructure...	68
Figure VI.2 Left: Scheme of a polycrystalline diamond based heterostructure with embedded IDT within the Sc _{0.43} Al _{0.57} N thin film. Centre and right: SEM images of an embedded IDT in a 300 nm thick Sc _{0.43} Al _{0.57} N thin film.....	69

List of tables

Table II.1 (Adapted from PAPER 1) Sputtering parameters employed for the synthesis of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films.....	17
Table II.2 Sputtering parameters employed for the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.....	17
Table II.3 Sputtering parameters employed for the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.....	18
Table IV.1 Conditions employed for the synthesis of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ (left) and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ (right) thin films.....	45
Table IV.2 Effective acoustic velocity and effective K_{eff}^2 coefficient obtained from the series and parallel resonances of the resonance frequency modes generated on the SAW devices with three different $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ heterostructures.....	59
Table IV.3 -3 dB bandwidth and centre of frequency f_c of the Rayleigh and Sezawa modes in SAW filters fabricated on the two different $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ heterostructures.....	62

Agradecimientos

Me gustaría agradecer a Gonzalo Fuentes por haber apostado por mí como su estudiante de doctorado. Muchas gracias por tus innumerables consejos y motivación para mejorar en todos los trabajos y actividades que hemos llevado a cabo durante estos años.

I am extremely thankful to everyone who has participated directly in this work. Zhuohui Chen from Huawei, for your simulations and designs. Oliver A. Williams from Cardiff University, for supplying the diamond substrates. Likewise the group from IAF who carried out several measurements, and explicitly Agnė Žukauskaitė and Yuan Lu, thank you for the fruitful discussions.

Thank you, professor Ruyen Ro for accepting and teaching me during my stay in Taiwan.

Gracias a todos los miembros del ISOM, sin el trabajo de cada uno de vosotros todo habría resultado imposible. Fernando Contreras y Óscar García, sin vuestro trabajo no tendríamos de los equipos funcionando como deben. Ana Pérez y Maika; empleasteis innumerables horas enseñándome cómo manejar por el laboratorio. Steven y Ana, cuidándonos y aconsejando en el despacho. Antonio Utrilla, cómo valoro todos y cada uno de tus consejos. A mis compañeros del C-206, Alejandro, Eduardo y Miguel siempre dispuestos a ayudar. David Castilla, qué suerte he tenido de poder aprender de cada paso que has dado desarrollando tus dispositivos. Lazar Stanojevic, por ponerme al día con lo que ocurría en el mundo. Manuel Abuín, muchas gracias por toda tu paciencia, enseñanzas y consejos. David López, sin tu trabajo y dedicación no habría podido ser capaz de salir del pozo experimental en el que me encontraba. Alberto Bosca, siempre buscando el lado positivo de las cosas. Miguel Montes, has sabido dar con la tecla para transmitir cada vivencia y consejo, muchas gracias. Antonio Ladrón, te nos fuiste pero volverás por todo lo alto. Rajveer Fandan, Julen Tamayo, Alicia Gonzalo y Amalia Fernando. Habéis sabido aportar cada pequeño granito para mejorar cada día. Thank you, Raj, for every life-changing conversation we had. Julen, muchas gracias por estar atento al mínimo detalle, por cada charla y cada momento. Alicia Gonzalo, incluso con tus interminables archivos de datos siempre has sacado un segundo para animarme y aconsejarme, muchas gracias. Amalia, siempre con una sonrisa, dispuesta a escuchar y ayudar, ojalá siempre tengas un hueco en esa apretada agenda para tus amigos.

Gracias a la peña “los Trileros” de mi pueblo, Riópar. A muchos ya os lo he dicho, todo el bien que hacéis y la suerte que tengo de poder siempre desconectar del día a día y disfrutar con vosotros.

Gracias a mis amigos Kike, Lorena, Almu y Quílez, a pesar del poco tiempo que hemos podido coincidir, siempre sois capaces de hacerme sonreír y disfrutar, incluso en los peores días.

Gracias a mis amigos Carlos, Alberto y Miguel. Sabéis lo importantes que sois. Siempre estáis dispuestos a ayudar. Las diferentes situaciones que hemos vivido estos últimos años me hacen valorar cada momento con vosotros. Muchas gracias Miguel. Estos años has sabido estar aún en la distancia. Has aguantado chaparrones y has sabido siempre aportar aquello necesario para mejorar cada momento contigo.

Gracias Borja, amigo. Las circunstancias me piden escribirte libros enteros. No te haces a la idea de la suerte que tenemos los que te conocemos. Nos transmites una fortaleza y entereza que a todos los que te rodeamos inspira. Ya sabes 10, sigue buscando siempre mejorar.

Finalmente, me gustaría agradecer el apoyo incondicional que siempre he recibido por parte de mis padres y hermanas. Siempre habéis confiado en mí. Siempre me habéis impulsado para mejorar. Siempre me habéis indicado por dónde podía continuar o aportado el consejo necesario para poder adaptarme a cada situación. Gracias.

Gracias Beatriz, tú mejor que nadie sabe de todo lo vivido durante el desarrollo de esta tesis. Nada me hace más feliz que poder agradecerte todo lo que has aportado en mi vida. Te quiero.

Miguel Sinusía Lozano

Publications

PAPER 1 Synthesis of ScAlN thin films on Si (100) substrates at room temperature, A. Pérez-Campos, **M. Sinusía Lozano**, F. J. Garcia-Garcia, Z. Chen, G. F. Iriarte, *Microsyst. Technol.* **2018**, 24, 2711, <https://doi.org/10.1007/s00542-017-3660-x>

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PAPER 2 Piezoelectric characterization of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ layers on Si (001) substrates, **M. Sinusía Lozano**, A. Pérez-Campos, M. Reusch, L. Kirste, T. Fuchs, A. Žukauskaitė, Z. Chen, G. F. Iriarte, *Mater. Res. Express* **2018**, 5, 036407, <https://doi.org/10.1088/2053-1591/aab232>

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PAPER 3 Temperature characteristics of SAW resonators on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures, **M. Sinusía Lozano**, Z. Chen, O. A. Williams, G. F. Iriarte, *Smart Mater. Struct.* **2018**, 27, 075015, <https://doi.org/10.1088/1361-665X/aabca4>

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PAPER 4 Giant Reflection Coefficient on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ Polycrystalline-Diamond SAW Resonators, **M. Sinusía Lozano**, Z. Chen, O. A. Williams, G. F. Iriarte, *Phys. Status Solidi A*, <https://doi.org/10.1002/pssa.201900360>

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I Introduction

The development of 5G telecommunication technologies not only increases the front-end micro electromechanical systems (MEMS) market, but also elevates the technological requirements for this type of devices. 5G technology is planned to support faster mobile broadband speeds completely enabling the internet of things (IoT). However, the success of this technology is highly dependent on government regulations in regards of the available radiofrequency spectrum. To deliver a widespread coverage, the 5G spectrum aims three different frequency spectrums: sub 1 GHz, 1-6 GHz and above 6 GHz [1]. For RF filters in particular, several requirements stand out such as the increase of power handling, miniaturization, manufacturing cost, thermal stability and the increment of bandwidth and working frequencies [2][3].

Multiple industries employ MEMS devices in different applications such as sensors, filters or accelerometers. Specifically, surface acoustic wave (SAW) devices are a promising MEMS solution for the technological constraints set by the developing 5G technology. The versatility of SAW devices to further develop their performances have attracted the industry and the academia since the first SAW device was commercialized in the mid-1960's. In this regard, multiple device designs have been proposed, new fabrication routes developed, and the use of novel piezoelectric thin films and substrates expands the utilisation scope of these devices.

Sensory applications employ SAW devices due to their high stability in harsh environments and measurement resolution. Furthermore, lab-on-a-chip applications have been recently developed with nanomolar resolution [4][5]. On the other hand, novel applications leverage the strain field created by the propagating SAW. For example, 2D plasmonic devices exploit it for coupling electrons and light, whereas spintronic applications couple electron spin and SAW reducing the current needed for spin rotation [6][7].

Concerning the developing of new piezoelectric thin films, the scandium aluminum nitride (ScAlN) compound is a successful example of the research advances carried out during the last decade. In 2009, Akiyama *et al.* reported the increase of the aluminum nitride (AlN) piezoelectric d_{33} constant by introducing scandium (Sc) within the piezoelectric AlN phase [8]. Since then, several groups have reported AlN based ternary and quaternary compounds with augmented piezoelectric responses as compared with the pristine AlN [9][10][11][12].

This thesis is devoted to the study of the ScAlN compound, from its synthesis to its feasibility to be employed in SAW devices. In this chapter the piezoelectric effect and its leverage in generating and propagating the SAW is covered. The last section in the chapter summarizes the work carried out during this thesis and how this document is organized.

1 Piezoelectricity

First discovered by the Curie brothers in 1880 [13], the direct piezoelectric effect relates the generation of an electrical moment when a strain is applied to the piezoelectric material. Inversely,

Chapter I

the converse piezoelectric effect, mathematically predicted by Lippman [14] and experimentally verified by the Curies in 1881 [15], occurs when the crystal is strained by the application of an electric field.

The equations for direct (I.1) and converse (I.2) piezoelectric effect state the relationship between the polarization vector (P_i), stress tensor (σ_{jk}), strain tensor (ϵ_{jk}), electric field vector (E_i) and the piezoelectric constants (d_{ijk})¹ [16].

$$P_i = d_{ijk} \sigma_{jk} \quad (\text{I.1})$$

$$\epsilon_{jk} = d_{ijk} E_i \quad (\text{I.2})$$

Piezoelectricity can only occur when a crystal class lacks a center of symmetry. If the direct piezoelectric effect is considered, the application of strain changes the charge distribution on the lattice resulting in a net electrical polarization of the crystal.

20 out of the 32 crystallographic point groups (also referred to as classes) in which crystalline materials are classified present enough asymmetry to exhibit piezoelectricity. In the particular case of the ScAlN compound, the *hexagonal phase class 6mm* presents piezoelectricity. Applying the corresponding symmetry conditions (I.3) reduces the 18 independent elements d_{ij} of the piezoelectric matrix to 5, as represented in matrix (I.4) [16].

$$\begin{array}{c} d_{11} = d_{16} = -d_{12} = d_{21} = d_{26} = d_{22} = 0 \\ \hline d_{15} = d_{24} \qquad \qquad \qquad d_{14} = -d_{25} \qquad \qquad \qquad d_{31} = d_{32} \\ \hline d_{13} = d_{23} = d_{36} = d_{35} = d_{34} = 0 \\ \hline \text{ScAlN case (class 6 mm):} \qquad \qquad \qquad d_{14} = -d_{25} = 0 \end{array} \quad (\text{I.3})$$

In the case of the ScAlN compound, the hexagonal phase class 6mm presents piezoelectricity. If this phase is synthesized perpendicular to the substrate surface, the piezoelectric response is highly affected by the piezoelectric d_{33} constant.

$$\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \quad (\text{I.4})$$

A parameter inherent to any piezoelectric material and of primary interest in electroacoustic applications is the piezoelectric coupling coefficient (K_{ij}^2) (I.5). This parameter measures the ability of the piezoelectric material to convert the electrical energy into mechanical energy and vice-versa. It is related to the piezoelectric stress matrix (e_{ij}), the stiffness matrix (c_{ij}) and the relative dielectric matrix (ϵ_{ij}) of the piezoelectric thin film [17]. The superscripts ^E and ^S denote that these constants

¹ Matrix notation is employed because of its compactness:

$$\begin{array}{c} \text{Tensor notation} \\ \left[\begin{array}{cccccc} d_{111} & d_{122} & d_{133} & d_{123} & d_{113} & d_{112} \\ d_{211} & d_{222} & d_{233} & d_{223} & d_{213} & d_{212} \\ d_{311} & d_{322} & d_{333} & d_{323} & d_{313} & d_{312} \end{array} \right] \end{array} \quad \text{to} \quad \begin{array}{c} \text{Matrix notation} \\ \left[\begin{array}{cccccc} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{array} \right] \end{array}$$

must be measured with the piezoelectric thin film grounded (zero electric field) and in a zero-stress condition respectively.

$$K_{ij}^2 = \frac{e_{ij}^2}{c_{ij}^E \epsilon_{ij}^S} \quad (I.5)$$

2 Surface acoustic waves (SAW) devices

An elastic (acoustic) wave involves changes (strains) in the relative positions of the atoms within a solid material [18]. These strains, developed as the wave propagates, cause internal stresses due to the internal forces generated when the atoms (or particles) tend towards their initial equilibrium positions [19]. In regard to the particle displacement, acoustic waves can be divided in longitudinal and shear waves. In a longitudinal wave the particle displacement is parallel to the propagation direction whereas in a shear wave such displacement is normal to the propagation direction.

The velocity of the acoustic wave (V_{ij}) is determined by the stiffness (c_{ij}) and density of the media (ρ_{ij}) in which the wave propagates (I.6). For that reason, large acoustic velocities are achievable only in materials that present high stiffness and low densities, such as diamond.

$$V_{ij} = \sqrt{\frac{c_{ij}}{\rho_{ij}}} \quad (I.6)$$

The so called Rayleigh wave, first predicted in 1885 by Lord Rayleigh [20], was the first demonstration of a surface acoustic wave. The boundary condition for their propagation is that there is no traction on the surface, and for that it has longitudinal and shear wave components. The Rayleigh wave propagates with an elliptical motion of the atoms within the material surface, whose amplitude decays exponentially in the direction normal to the surface. It is generally accepted that a surface acoustic wave is completely extinguished in a depth of three times its wavelength.

Love and Sezawa waves, named after A. E. H. Love [21] and K. Sezawa [22], are surface acoustic waves reported afterwards. Love waves are purely shear waves with a particle displacement normal to the propagation direction. Pure shear waves are incompatible with the condition of an unstressed surface. For that reason, Love waves require of a layered structure and have a velocity of propagation that varies with depth. Thus, Love waves are generally employed in sensory applications in liquid media because their pure shear component is not attenuated in its contact with the fluid.

Sezawa waves are usually referred to as the first Rayleigh mode. The elliptical motion of the atoms in a Sezawa wave is opposite to that of the Rayleigh wave around the ellipse. Sezawa waves require a minimum layer thickness of a media with a lower acoustic velocity than the substrate below to be generated and propagated unattenuated [23]. These waves propagate at larger velocities than the fundamental Rayleigh mode and thus, depending on the structure in which it propagates, provide large capabilities when used in electroacoustic devices.

Heterostructures, a stacking of different material layers, are employed in electroacoustic applications when working with polycrystalline piezoelectric thin films. The basic heterostructure comprises a substrate on which the piezoelectric layer is deposited. In thin film based SAW devices, the most common design set an interdigital transducer (IDT) on top of the heterostructure to generate and detect the surface acoustic wave. In order to confine the acoustic wave, reflectors can be placed at the end of the IDT (Figure I.1). An IDT consists of strips (fingers) in a periodic comb-like structure connected to bus bars. When these bus bars are connected to a RF source, the electrical signal induces mechanical strains within the piezoelectric thin film radiating a surface acoustic wave from the IDT. Thus, the designed IDT wavelength sets the wavelength of the propagating SAW [19].

There are several IDT configurations, but the single-electrode-type IDT is widely employed because of its design simplicity. In the single-electrode IDT type, the pitch formed by two adjacent strips with opposite polarity (a finger pair) is half the designed wavelength of the IDT. Several aspects must be carefully considered when designing the IDT such as the aperture (distance between the finger ends with opposite sign), the finger width or the metallization ratio (which is the ratio between the finger width and free space within the pitch). Further details about the IDT design and the influence of these definitions can be found in the comprehensive book of K. Hashimoto [19].

As it has been previously mentioned, reflectors placed at the end of the active region confine the SAW within the IDT. There are several reflector designs, ranging from a single metal strip, floating strips or the more sophisticated designs such as grounded strips patterned with the IDT wavelength. Since a surface acoustic wave is confined within a depth of 3λ , the SAW acoustic velocity largely depends on the heterostructure materials through which the wave propagates. Each particular IDT/heterostructure combination can be studied using different mathematical models [23]. These can be divided in phenomenological methods such as the P-method or the coupling of modes (COM) method [24], and numerical methods which extract information from the device directly from the material constants such as finite element methods (FEM) and/or solving the Green's function [19].

Figure I.1 Scheme of a single-electrode interdigital transducer (IDT) with reflectors at both ends

The results of these simulations are graphically represented in “dispersion curves” which are normalized to the ratio between the piezoelectric thin film thickness and the designed IDT wavelength. Focusing on the ScAlN compound, Hashimoto *et al.* have reported several dispersion curves for different stacking layer configurations and substrates such as diamond and 6H-SiC substrates [25][26][27].

A one port resonator consists of one IDT and depending on the application, a set of reflectors to confine the generated standing wave (Figure I.2). The basic structure without weighting the IDT response is the comb structure previously commented (Figure I.2 a). However, there are several weighting designs employed to modify the response of the IDT. Double electrode transducers (Figure I.2 b) are employed to reduce the distortion originated by the weak reflections experienced by the generated SAW in each IDT finger. When these weak reflections add in phase, they cause the distortion of the signal degrading the resonator response [28]. The apodization of the IDT design (Figure I.2 c) is largely employed because of the control it offers on the weighting as the SAW excitation is proportional to the overlap between consecutive IDT fingers [19]. The weight weighting (Figure I.2 d), in which the pitch remains constant whereas the metallization ratio varies,

is difficult to implement because of the complicated relationship between SAW excitation and the finger width variations [19].

Figure I.2 One-port resonator configurations a) IDT design without weighting; b) IDT design with a double electrode transducer weighting; c) IDT design with apodized weighting; d) IDT design with weight weighting

Regarding the SAW filter technology, there are two different device designs: those leveraging the delay line characteristics of the propagating SAW and those using the impedance characteristics of several connected resonators. Double mode SAW (DMS) filter designs use two resonant modes acoustically coupled in the longitudinal direction (Figure I.3 a). The DMS filter bandwidth can be adjusted controlling the piezoelectric thin film thickness, and the IDT design. SAW based radiofrequency identification (RFID) tags use the propagation characteristics of the SAW providing device robustness and an infinite number of geometries to use (Figure I.3 b) [29]. On the other hand, ladder and bridge filter configurations connect several one port resonators with slightly different resonance frequencies (Figure I.3 c, d). These filter configurations provide large bandwidths, low insertion losses and high power handling capabilities, which can be achieved by adjusting independently the impedance characteristics of each resonator [30].

Figure I.3 SAW based filter designs a) Double mode SAW (DMS) filter; b) Z-path SAW tag; Scheme of a c) ladder d) bridge electrical connection

3 Scandium aluminum nitride (ScAlN) compound

Undoped AlN has been extensively employed in a large variety of electroacoustic thin film applications. If its properties are compared to those of other compounds that are usually employed in these applications, such as ZnO, AlN outstands because of its high thermal and chemical stability, high stiffness and its high piezoelectric d_{33} constant [31][32][33].

The $\text{Sc}_x\text{Al}_{1-x}\text{N}$ compound is the result of alloying AlN with Sc. This ternary system has been extensively studied since 2009, when Akiyama *et al.* experimentally demonstrated a gradual increase of the piezoelectric d_{33} constant with the Sc content within the $\text{Sc}_x\text{Al}_{1-x}\text{N}$ compound [8]. The maximum increase reported (500 %) corresponds to a $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ composition. The former has been explained via *ab initio* simulations [34][35][36][37]. The reason behind the increase of the piezoelectric constant is the substitution of the Al atoms by Sc atoms within the wurtzite phase of AlN. Such an inclusion modifies the lattice constants and bond angles of the pristine wurtzite AlN phase which are responsible, together with a decrease of the covalent bond strength, of the influence of the Sc concentration on the overall piezoelectric response of the ScAlN compound [38]. It has also been shown how an increasing Sc content is not only responsible for a higher piezoelectric constant, but it also reduces the compound elastic constant and alters optoelectronic properties such as the bandgap [39][40][41][42].

However, the progressive d_{33} increase is halted because of the competitive synthesis of a rock salt (non-piezoelectric) phase, which is the most energetically favoured phase for Sc contents above ~55 % [34][35][36]. On the other hand, the mixing enthalpy calculations show that the synthesis of the metastable wurtzite ScAlN thin films is experimentally achievable in non-thermodynamic equilibrium conditions like the ones provided by the reactive sputtering technique [34][35][36].

Since Akiyama *et al.* published their work in 2009, multiple research groups with a large expertise in the synthesis of AlN by reactive sputtering turned their research towards the promising ScAlN compound. Therefore, process and synthesis parameters that were studied and optimized during decades for the synthesis of highly c-axis oriented AlN thin films were taken as a starting point to study the reactive sputtering synthesis of ScAlN thin films. Process parameters encompass those constrained by the geometry and configuration of the equipment, such as the target to substrate distance, the target size or the possibility to use either a single alloy target or the co-sputtering technique which employs two independent pure metallic scandium (Sc) and aluminum (Al) targets [43][44]. Aiming to control the ScAlN thin film composition, several research groups proposed the use of single aluminum targets with embedded Sc ingots [45]. Temperature, process pressure and discharge power are, among others, usually studied and addressed to synthesize highly c-axis oriented ScAlN thin films [36][46]. Additionally, an AlN seed layer can be employed to further promote the c-axis orientation [47]. Originally, seed layers matching the three-fold symmetry of the wurtzite AlN were considered. Examples of these are hexagonal structures such as titanium (Ti) (002) or cubic structures like platinum (Pt) (111), gold (Au) (111), Al (111) or titanium nitride (TiN) (111) [48][49][50][51]. However, the need of electrode materials for several electroacoustic applications (for example for thin film bulk acoustic resonators (FBAR)) and compatibility with silicon technologies forced the synthesis of c-axis oriented ScAlN and AlN thin films on metallic layers without resembling the symmetry of the c-axis ScAlN orientation, as for example cubic textured layers of molybdenum (Mo) (110) or chromium (Cr) (110) [52]. However, independently of using any seed layer, successful examples of highly c-axis oriented AlN and ScAlN thin films have been reported on a large variety of substrates such as diamond (single and polycrystalline), silicon, silicon oxide and sapphire (Al_2O_3) (0001) [53][54][55].

The synthesis of ScAlN has also been reported to be feasible using other techniques such as molecular beam epitaxy (MBE) [56], hydride vapour phase epitaxy (HVPE) [57] or physical vapor transport (PVT) [58] aiming to include the ScAlN compound in optoelectronic applications.

4 Aim of the thesis

The main objective of this thesis is the study of the ScAlN compound and its feasibility for being used in SAW devices for 5G applications. These devices should meet several 5G technological requirements such as the power handling capabilities, temperature stability and 5G working frequencies and bandwidths. In order to meet these constraints, the proposed heterostructure comprises a ScAlN thin film on a diamond substrate.

The power handling capability is tackled from the heterostructure design and the thermal conductivity of the materials it comprises. In high power applications, the IDT degrades rapidly by the stress migration of the metal induced by the temperature rise and the displacements of the surface caused by the propagating SAW. The use of diamond as a substrate aids in spreading the heat generated by joule effect in the IDT metal, whereas its high elastic constants reduce the displacement caused by the SAW.

By using high acoustic velocity materials such as diamond, higher resonance frequencies are achievable without demanding large nanolithography resolutions. The maximum achievable bandwidth is set by the effective electromechanical coupling coefficient of the heterostructure. Thus, the large piezoelectric d_{33} constant and thus the K^2 reported for the ScAlN compound motivates its use to attain wider bandwidths.

The temperature coefficient of frequency (TCF) provides an overall measure of the device thermal stability. The different materials employed to fabricate the device do not only have to sustain their properties under extreme temperatures but to handle wide temperature ranges. In order to cope with this thermal constraint, the materials within the device can be selected so that their thermal

expansion coefficients counter the temperature induced strains generated in each layer providing the required temperature stability.

Taking all this into account, different works were carried out focusing on partial objectives which are chronologically listed below:

- A. Synthesis of the ScAlN compound with a $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target.
 - a. Study of the sputtering process to promote the synthesis of highly c-axis oriented $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films.
 - b. Structural, compositional and piezoelectric characterization of the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films, focusing on the influence that the synthesis parameters have on the piezoelectric response of the thin film.
 - c. Synthesis of highly c-axis oriented $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films on high acoustic substrates (polycrystalline diamond).
- B. Fabrication and characterisation of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ based SAW devices.
 - a. Optimisation of the nanolithography and lift-off processes.
 - b. Electrical characterization of SAW devices.
 - c. Curve fitting simulation of the electrical response of the devices.
- C. Synthesis of the ScAlN compounds with a $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target.
 - a. Study of the sputtering process to promote the synthesis of highly c-axis oriented $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.
 - b. Structural, compositional and piezoelectric characterization of the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films, focusing on the influence that the synthesis parameters have on the piezoelectric response of the thin film.
 - c. Synthesis of highly c-axis oriented $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films on high acoustic substrates (polycrystalline and single crystal diamond).
- D. Fabrication and characterisation of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ based SAW devices.
 - a. Electrical characterization of SAW devices.
 - b. Curve fitting simulation of the electrical response of the devices.
 - c. Improve the design and device performance.

5 Structure of the thesis

The second chapter describes the experimental techniques employed during this thesis. Among others, the reactive sputtering technique is addressed, discussing the influence of the different sputtering parameters on the process. The home-built sputtering system is described here, together with the conditioning procedure followed prior to each synthesis process. Finally, the different characterization techniques are introduced and literature for further reading is recommended for each technique.

Chapter III focuses on the studies carried out to promote the synthesis of highly c-axis oriented ScAlN thin films. The influence on the texture and piezoelectric response of the different parameters employed in the synthesis is shown. This influence is presented independently for the compounds synthesized using the $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy targets.

The fourth chapter focuses on the electroacoustic characteristics of several SAW devices fabricated using different heterostructures. SAW resonators fabricated on ScAlN thin films synthesized with both alloy targets are presented. Finally, the electrical characterization of ScAlN based RF SAW filters is addressed.

Chapter V summarizes the work presented in this thesis highlighting the most relevant results, whereas in the sixth chapter different work routes to be studied in the future are introduced. For example, increasing the device power handling by embedding the IDT on the piezoelectric thin film layer, or increasing the working frequencies reducing the thin film thickness of the ScAlN thin film leveraging the extremely high acoustic velocity of substrates such as diamond.

II Experimental

This chapter focuses on the synthesis, fabrication and different characterization techniques employed during this thesis. Firstly, the reactive sputtering technique is presented. Afterwards, the home-built reactive sputtering system that has been used to synthesize the ScAlN thin films is detailed, together with the basics of the nanolithography and the subsequent lift-off processes employed to fabricate the SAW devices. The physics behind the different characterization techniques is briefly addressed and literature on each technique is proposed for further reading. Finally, the electrical characterization of the fabricated SAW devices as well as the equivalent circuit model employed to simulate the device performance are presented.

1 Reactive sputtering

This thesis studies the synthesis of highly c-axis oriented ScAlN thin films by reactive sputtering. This section focuses on the reactive sputtering process and how the most relevant parameters influence the synthesis process.

1.1 Background

The sputtering technique is based in the deposition of particles ejected from a material source (target). An electric potential difference between the anode (chamber surface) and the cathode (target) ionizes the gases (plasma) within the synthesis chamber. The ionized species impinge into the target with enough energy to overcome the binding energy of its surface atoms. These collisions cause not only the sputtering of the target surface species, but also the emission of other particles such as secondary electrons and neutrals. The secondary electrons aid in sustaining the plasma as they collide with the species in the gas phase, providing additional energy for their ionization.

However, in order to efficiently leverage the energy of the emitted secondary electrons, magnetrons are employed to create a magnetic field which confines ions and electrons close to the target surface. This increases the probability of collisions, yielding in a higher ionization probability and denser plasmas. Because of the former, more stable plasma conditions (no arcing) are sustained at higher discharge powers and lower process pressures which is reflected in the overall energy of the sputtered particles arriving on the substrate surface. Experimentally, a special characteristic of the plasma confinement caused by the magnetron is the generation of a “race-track” on the target surface. It follows the magnetron geometry (and thus the magnetic field) and is an area at the target surface where it is more eroded.

Chapter II

The race-track in new targets must be created warily, controlling the process parameters, mainly the process pressure and the discharge power, in order to avoid surface damage or even breakage of the target.

In the case of dielectric compounds, as it is the case with most of oxides and nitrides synthesized by reactive sputtering, pulsed DC or RF discharges should be employed for discharging the target surface of positive ions that would cause arcing [59]. The plasma stability of the former improves considerably because of its steeper transition between positive and negative pulses. When using pulsed DC, the pulse width and duty cycle (time ratio between positive and negative pulses) have to be adjusted in order to obtain stable and arc-free plasma conditions [60].

1.2 Reactive sputtering

Reactive sputtering is based in the synthesis of the compound that the reaction between the target material and the reactive gas creates. This technology is extensively employed in thin film coating and semiconductor industries. Modelling the process parameters and the thin film outcome of the reactive sputtering process aids in implementing the technology in processes relevant for these industries. Because of its simplicity and reliability, “Berg’s model” is extensively referred to [61]. Several research groups have further developed Berg’s model including specific process parameters such as ion implantation [62][63], sticking coefficients of the chemical reactions [64] or the discharge voltage behaviour [65].

When the reactive gas (N_2 for instance) is introduced into the synthesis chamber, it reacts with the particles in the gas phase covering the target surface (as well as any other surfaces within the synthesis chamber) with a nitride compound. The compound coverage of the target surface causes a hysteresis effect in the deposition rate, which is characteristic of the reactive sputtering process [61]. The compound formation on the target, which is counter by the ion bombardment, determines the two differentiated steady states of the aforementioned hysteresis effect, known as metallic and compound modes.

1.3 Synthesis of c-axis $ScAlN$ thin films by reactive magnetron sputtering

In the literature, several target configurations have been employed for the synthesis of $ScAlN$ thin films by reactive sputtering (Figure II.1). The co-sputtering technique can tune the thin film composition by controlling the discharge power supplied to two independent pure metallic, aluminum (Al) and scandium (Sc), targets (Figure II.1 Left) [66]. On the other hand, when a single alloy target configuration is employed a Sc deficiency within the synthesized $Sc_xAl_{1-x}N$ thin film, as compared to the Sc_xAl_{1-x} alloy target, has been reported [67][68][69]. Furthermore, $ScAlN$ thin films with Sc contents up to 15 % have been reported when scandium inlets are introduced into the race track of the aluminum target (Figure II.1 Right) [70][71].

Figure II.1 Different sputtering target configurations. Left: Two targets for a co-sputtering configuration. Right: Aluminum single target with scandium inlets in the race-track

In this thesis, $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films have been synthesized using the reactive sputtering technique from two different alloy targets, $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$ respectively, with no intentional substrate heating. The synthesis parameters have been studied in order to promote highly c-axis oriented ScAlN thin films. Process pressure, temperature, discharge power, radial position of the substrate with respect to the target, target-to-substrate distance or deposition rate are among the sputtering process parameters that have a major influence in the microstructure of the synthesized thin film. For that reason, their influence is briefly addressed in section II.1.4.

However, the energy of the arriving species on the substrate surface must be addressed before. Resuming the models of nucleation and growth of thin films on surfaces, there are several major steps in the thin film growing process: adsorption, nucleation, nuclei coalescence and thin film growth [72][73]. The thin film nucleation process starts when an incident atom adsorbs in the substrate surface (adatom). The adatom diffuses towards an energetically favourable position creating nucleation centres. When these nucleation centres reach a critical size, they coalesce, commencing the growth of the thin film. Therefore, the mobility of the adatoms (which is determined by their energy as they adsorb on the substrate surface) strongly influences the microstructure evolution of the growing thin film. Internal stresses, voids and defects can be cured to a large extent by controlling the synthesis conditions.

1.4 Influence of the sputtering parameters

Deposition rate

Every sputtering parameter commented in the following influence on a large extent the amount of sputtered particles and thus the deposition rate. When the deposition rate is considerably large, the new arriving species set the position of the diffusing adatoms in non-energetically favourable positions creating defects within the thin film. On the contrary, low deposition rates may cause an insufficient flow of particles to the substrate surface and consequently (especially at low synthesis temperatures) the adatom energy might not be enough to diffuse resulting in a deterioration of the synthesized thin film microstructure.

Pressure

Resuming the kinetic theory of gases, pressure and temperature are the major process parameters that control the mean free path between collisions (λ_{mfp}) of the particles in the gas phase [72]. This is of a significance in several steps of the sputtering process. The process pressure (λ_{mfp}) determines the amount of the collisions experienced by the ejected particles in their way through the gas phase.

The density of ionized plasma species will be larger at higher process pressures, and hence they will undergo many more collisions in their travelling towards the target. Thus, at high process pressures (when there is a considerable larger amount of collisions), the particles in the gas phase are “thermalized” as the ejected particles lose a portion of their kinetic energy in each collision [74].

Discharge power

The pulsed DC plasma generator used in this thesis powers the magnetron with a constant power control. It therefore adjusts the discharge voltage and current according to the plasma impedance.

The discharge power provides the ionization energy in the plasma and thus the kinetic energy of the impinging ions on the target surface. The energy of these ions is transferred via an elastic collision to the knocked-out atoms which travel through the gas phase. Once the substrate is reached, a portion of these travelling particles adsorbs on its surface, and the thin film growth begins. Therefore, the discharge power largely influences the kinetic energy that the adatoms need to diffuse on the substrate surface and initiate the thin film growth process.

A particular consideration should be taken when the kinetic energy of the arriving particles is large. In that case, the adatoms can be knocked-out from the substrate surface deteriorating the microstructure of the thin film by creating defects within it.

Temperature

Diffusion is a temperature-controlled parameter. Therefore, because of the large diffusion lengths at high synthesis temperatures, the growing thin films can cure voids and release stresses resulting in a defect-free thin film. However, in this thesis the synthesis is carried out with no intentional substrate heating, and therefore the adatom energy is provided by the kinetic energy of the arriving species on the substrate.

Substrate

The growing thin film resembles the lattice of the substrate in the first layers. The former has several implications at the lattice and microstructure level. If there is a large lattice mismatch, the first layers of the thin film accommodate their lattice parameter to that of the substrate, resulting in highly strained layers. This strain is released as the thin film grows; however, it may cause thin film peeling-off in stacked structures. The growing thin film resembles the substrate surface, therefore large substrate surface roughness complicates the ordering of the thin film grains resulting in thin films with disoriented grains or even amorphous thin films [50].

Dust particles and grease in the substrate surface do not only imply impurities within the thin films. They act as recombination centres, places where the adatoms conglomerate causing disordered structures. Since most of the deposition processes bake-out the substrate surface prior the deposition, they presume its cleanliness. However, in this thesis there is no substrate heating, which implies a thorough cleaning via a two-solvent method employing acetone at 60 °C, a sonication bath of methanol at room temperature and blown dry the substrate surface with N₂.

Gas admixture ratio ($Ar/(Ar+N_2)$)

In order to facilitate the ionization process, argon (Ar) is usually introduced with the reactive gas into the synthesis chamber. This not only eases the plasma ignition needing lower discharge voltages but increases the sputtering yield of the process. A larger mass increases the collision probability with the atoms on the target alloy surface whereas the kinetic energy, which is directly related to the mass, provides the energy in the collision. In the case of the Ar and N₂ gas admixture, the Ar⁺ ion mass is larger than the mass of an ionized N₂⁺ molecule and thus the sputtering yield is larger with larger Ar contents within the gas admixture of the synthesis.

Target-to-substrate distance

The thermalization of the sputtered particles determine the energy of the adatoms to diffuse. Therefore, the target-to-substrate distance is strongly related to the process pressure and thus to the mean free path between collisions (λ_{mfp}) and can be employed for controlling the amount of arriving particles to the substrate surface (deposition rate) and their energy.

Experimentally this distance is constrained by the synthesis chamber geometries, target size, and the relative positions of target, shutter and substrate holder.

Radial target-to-substrate position

The sputtering process follow a cosine distribution which cause spatial and energetic distributions of sputtered atoms. This is of significance in the synthesis from compound targets since compositional gradients and different deposition rates occur depending on the position at which the substrate is located with respect to the target centre [75]. In this thesis, the syntheses are carried out placing the substrate holder below the target alloy centre.

2 Synthesis and device fabrication

2.1 Home-built reactive sputtering system

The ScAlN thin films were synthesized using a home-built reactive sputtering system. It consists of two differentiated chambers, namely the load lock chamber and the synthesis one (Figure II.2). This configuration reduces the number of impurities in the thin films since the synthesis chamber is sustained at a base pressure below 9×10^{-5} Pa.

The magnetron is powered using an ENI RPG50 asymmetric bipolar-pulsed DC generator. The DC generator applies 250 kHz and in order to obtain stable plasma conditions the pulse width is set to 1616 ns. The N7.0 purity process gases namely nitrogen (N_2) and argon (Ar), are introduced into the synthesis chamber via 100 sccm mass flow controllers. The process pressure is controlled via a butterfly valve.

The (101.6 mm in diameter) alloy target is water-cooled in the backside. The normal target-substrate distance is set to 45 mm, with the substrate holder positioned below the target center. The temperature during the synthesis is monitored through a type K thermocouple placed below the substrate holder. Due to the aforementioned influence of the thin film composition on the compound piezoelectric constant, two alloy targets with different compositions, namely $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$, have been employed.

Figure II.2 Home-built sputtering system employed for the synthesis of ScAlN thin films

2.2 Synthesis conditions

Because of the variation of the piezoelectric response with the scandium content within the $\text{Sc}_x\text{Al}_{1-x}\text{N}$ compound, during this thesis the thin films have been synthesized using two alloy targets with different compositions, namely $\text{Sc}_{0.40}\text{Al}_{0.60}$ and $\text{Sc}_{0.60}\text{Al}_{0.40}$, have been employed. The $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target was the first employed to synthesize highly c-axis $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films (Table II.1).

Table II.1 (Adapted from PAPER 1²) Sputtering parameters employed for the synthesis of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films

Discharge power (W)	300, 500, 700, 900
Process pressure (Pa)	0.53, 0.80, 1.06
$\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)} (\%)$	25
Total gas flow $(\text{Ar} + \text{N}_2)$ (sccm)	12
Target-to-substrate distance (mm)	45
Initial substrate temperature (K)	298
Target alloy	$\text{Sc}_{0.40}\text{Al}_{0.60}$

When the $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target was retrieved from the sputtering system, its surface presented cracks because of the heat generated during the synthesis. For that reason, several preventive measures were taken both in the sputtering system running with the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target: the water-cooling system was revised, and a new magnetron installed, which increased the magnetic field within the plasma.

Regarding the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target, an indium bonded 3 mm thick copper backing plate was employed for heat dissipation (to avoid the target cracking). However, at discharge powers larger

² Adapted by permission from Springer Nature Customer Service Centre GmbH: Springer Nature, Microsystem Technologies, Synthesis of ScAlN thin films on Si (100) substrates at room temperature, A. Pérez-Campos, M. Sinusía Lozano, F. Javier Garcia-Garcia et al, 4661280082424, 2017

than 500 W, the indium melted causing the electrical short-circuit of the system. Furthermore, the copper plate attenuates the magnetic field within the plasma, and that together with the new magnetron, modified the overall impedance characteristics of the plasma. Thus, whereas with the $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target discharge powers up to 900 W and process pressures of 0.53 Pa were employed (Table II.1), when the syntheses were carried out using the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target, the compromise of using a maximum discharge power of 500 W was used. At 500W, the new magnetron aided in sustaining stable plasma conditions (no-arcing) down to a process pressure of 0.33 Pa (Table II.2).

Table II.2 Sputtering parameters employed for the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films

Discharge power (W)	300, 500
Process pressure (Pa)	0.40, 0.66, 0.93; only for 500 W: 0.33
$\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)} (\%)$	25
Total gas flow $(\text{Ar} + \text{N}_2)$ (sccm)	12
Target-to-substrate distance (mm)	45
Substrate temperature (K)	298
Target alloy	$\text{Sc}_{0.60}\text{Al}_{0.40}$

The syntheses to evaluate the influence of the gas admixture (ratio between the non-reactive gas and reactive gas flows employed during the synthesis $(\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)})$) on the thin film properties were carried out using the process parameters shown in Table II.3.

Table II.3 Sputtering parameters employed for the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films

Discharge power (W)	500
Process pressure (Pa)	0.40
$\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)} (\%)$	25, 37.5, 62.5, 75
Total gas flow $(\text{Ar} + \text{N}_2)$ (sccm)	12
Target-to-substrate distance (mm)	45
Substrate temperature (K)	298
Target alloy	$\text{Sc}_{0.60}\text{Al}_{0.40}$
$\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film thickness (nm)	300, 1000

2.3 Substrate cleaning

Several commercial substrates such as B-doped³ and As-doped⁴ Si (001) or thermally grown SiO_2 substrates⁵ have been employed to optimize the sputtering conditions necessary to synthesize highly c-axis oriented ScAlN thin films. The native oxide layer on the doped Si (001) substrates was not

3 Purchased from Crystal GmbH. <http://crystal-gmbh.com/> (Accessed on 12/09/2019). Datasheet: B-doped Si (001) resistivity 1-50 Ohm·cm.

4 Purchased from Silicon Quest International (Web page not available on 12/09/2019). Datasheet: As-doped Si (001) resistivity: 0.005 Ohm·cm.

removed. Finally, since the objective of this thesis is to fabricate ScAlN based SAW devices for 5G applications, diamond substrates have been employed. The polycrystalline diamond substrates have been provided by Prof. Oliver A. Williams, from Cardiff University. These polycrystalline diamond substrates have been synthesized by microwave plasma chemical vapour deposition (MPCVD) on top of 500 μm thick Si (001) substrates. Also, single crystal diamond (111) substrates⁶ were purchased to compare SAW propagation characteristics.

Before the synthesis, every substrate was cleaned using a two-solvent method which implied an acetone bath at 60 °C and a sonication bath at room temperature in methanol, both for 5 minutes. Afterwards the substrates were blown dry with N₂ before transferring them into the load-lock chamber.

2.4 Sputtering system conditioning

The synthesis chamber was subjected firstly to a pure argon atmosphere (30 sccm) 500 W discharge power at a process pressure of 1.33 Pa. Afterwards, nitrogen was introduced and the gas admixture ratio (Ar/(N₂+Ar)) adjusted maintaining the same discharge power and process pressure. Only afterwards, these two parameters were set to the values shown in Table II.1, Table II.2 or Table II.3. Each of these steps lasted 3 minutes. Finally, the substrates are transferred from the load lock chamber into the synthesis chamber and subjected to the final step of the conditioning process, with the specific synthesis conditions for 2 minutes with the shutter closed.

2.5 IDT fabrication

The IDTs were fabricated using a standard lift-off process (Figure II.3). A Crestec CABL-9500C e-beam lithography system was employed to pattern the different IDT designs.

Due to the insulating behaviour of the ScAlN thin film and the underlying polycrystalline diamond substrates, an organic anti-static layer (Espacer 300Z, Showa Denko) is spun on top of the e-beam resist to avoid charge accumulation during the e-beam lithography process.

⁵ Purchased from Cemat Silicon SA. (Web page not available on 12/09/2019). SiO₂ thermally grown on As-doped (111) Si. Datasheet Si resistivity: 0.001-0.005 Ohm·cm.

⁶ Purchased from EDP Corporation <http://www.d-edp.jp/en/index.html> (Accessed on 12/09/2019)

Figure II.3 Lift-off process employed for the fabrication a SAW device IDT on a ScAlN/polycrystalline diamond heterostructure

3 Structural characterization

3.1 X-ray diffraction (XRD)

The X-ray diffraction (XRD) technique is widely used for texture, phase and stress analysis of polycrystalline materials. This technique exploits the diffraction between an impinging X-ray wave and the periodic structure in which the crystals are ordered (Figure II.4 Left). Such reflection can be detected because of the comparable lengths of the crystal interatomic distances and the incident X-ray wavelength [76].

The maximum reflection occurs when the constructive interference described by the Bragg equation (II.1) is fulfilled (Figure II.4 Right). This equation relates the interplanar distance (d_{hkl}) between two adjacent planes described by Miller indices hkl , the X-ray wavelength (λ) and the incident and diffracted angle (θ).

$$n\lambda = 2 d_{hkl} \sin \theta \quad (\text{II.1})$$

The interplanar distance d_{hkl} depends on the crystal lattice. Thus, by introducing the interplanar distance of the lattice under study into the Bragg equation, the reflection angles of the scattered electrons can be obtained.

Figure II.4 Left: Scheme of the Bragg Brentano geometry for XRD analysis. Right: Visualization of the Bragg equation

The $\theta/2\theta$ scan

The $\theta/2\theta$ scan measures Bragg reflections and is extensively used in the study of polycrystalline thin films. In these scans, the angles of the incident and the reflected beams are equal thorough the scan. This condition is fulfilled by moving the detector in a velocity twice than the incident angle θ . Microstructural information can be derived from the position, shape, relative intensity etc., of the different reflections.

The ω scan or Rocking curve

The full width at half maximum (FWHM) of the ω scan or rocking curve provides a quantitative information of the degree of orientation that the thin film presents in the measured texture. In these scans, the detector is placed at the Bragg reflection angle (2θ) under study, whereas the sample is “rocked” around the angle at which the Bragg condition is fulfilled.

4 Surface characterization

4.1 Scanning electron microscopy (SEM)

The scanning electron microscopy (SEM) is widely employed because its variety of operation modes, nanoscale resolution and neglectable sample preparation. The most common mode of this technique relies on the detection of secondary electrons scattered from the sample surface after the electrons of a high energy electron beam interact with its atoms [77]. These secondary electrons scape the specimen surface with very low kinetic energies which are recorded by multiple solid-state detectors. X-rays or backscattered electrons may also be detected providing information on the sample's composition (Figure II.5 Left). The signal of these detectors is subsequently employed to determine the intensity (grey scale) of the pixel displayed on the computer screen.

Figure II.5 Scheme of several energies produced by the interaction of the focused electron beam. Secondary electrons are recorded by the SEM equipment, whereas the energy dispersive X-ray spectroscopy (XEDS) technique relies on detecting the characteristic X-rays

4.2 Atomic force microscopy (AFM)

The atomic force microscopy (AFM) lies within the broader family of techniques termed scanning probe microscopy (SPM) [78]. In this technique, the sample surface is probed with an extremely sharp tip, placed at the end of a cantilever, which allows the study of the surface topography at atomic scale (II.6). Using a reflected laser beam, a photodetector records the cantilever deflections caused by the attractive and repulsive forces between the tip and the sample surface. The root mean squared R_{RMS} (II.2) or the arithmetical mean deviation R_a (II.3) are calculated using the vertical distance from the mean line to the i^{th} data point (y_i). Both parameters are commonly used to indicate the roughness of the sample surface.

$$R_{RMS} = \sqrt{\frac{1}{n} \sum_{i=1}^n y_i^2} \quad (II.2)$$

$$R_a = \frac{1}{n} \sum_{i=1}^n |y_i| \quad (II.3)$$

Figure II.6 Scheme of the AFM equipment

5 Compositional characterization

Because of their inherent resolution, several techniques have been employed in this thesis to characterize the composition of the synthesized ScAlN thin films. Whereas several analysis techniques rely on the interaction between a focused high energy electron or ion beam with the atoms within the thin film, others detect the atoms sputtered from a small area. In this section, the working principles of each analysis technique are briefly described.

5.1 Energy dispersive X-ray spectroscopy (XEDS)⁷⁸

The energy dispersive X-ray spectroscopy (XEDS) is based in the detection of the characteristic X-ray photon emitted by a sample irradiated with a high energy electron beam. Because of the interaction between the arriving high energy electron and the sample nuclei, the atoms get ionized, causing the ejection of an inner shell electron. When an outer shell electron fills in that shell, the energy difference between the outer and inner shells is released in the form of an X-ray photon (Figure II.5). The energy of this X-ray photon is unique for every element, which allows the elemental or chemical characterization of the sample composition [77].

5.2 Secondary-ion mass spectrometry (SIMS)⁹

The secondary-ion mass spectrometry (SIMS) technique studies the energy of ejected secondary ions using a mass spectrometer (Figure II.7 Left). These secondary ions are emitted when focusing a high energy ion beam on the sample. High energy ions travel several monolayers across the thin film thickness, impinging with the atoms and exchanging energy causing a collision cascade (Figure II.7 Right). When some of these collisions return to the surface with enough energy to break the atomic bonds, material is ejected in the form of atoms. Some of these get ionized in the process of leaving the sample surface and detected [79]. This technique outstands because of its sensitivity ($\sim 10^{-4}$ at. %), and its capability of detecting, with few exceptions, all elements in the periodic table providing information of the layer thickness. However, for a quantitative chemical analysis the need of compositional standards is mandatory.

7 The XEDS measurements and analyses of [PAPER 1](#) were carried out using a Jeol JSM 7600F system, equipped with an INCA X-ray detector, at ICTS-Centro Nacional de Microscopía Electrónica, by F. Javier Garcia-Garcia.

8 The XEDS measurements and analyses of the $\text{Sc}_{0.47}\text{Al}_{0.53}\text{N}$ thin film have been carried out using a Zeiss Auriga Cross-beam FIB-SEM equipped with a Bruker Quantax EDX module, by the “Emerging materials department, piezoelectric group” at Fraunhofer Institute for Applied Solid State Physics IAF.

9 The SIMS measurements and analyses have been carried out using a Cameca 4500 quadrupole system, by the “Emerging materials department, piezoelectric group” at Fraunhofer Institute for Applied Solid State Physics IAF.

Figure II.7 Left: Scheme of the SIMS equipment. Right: Schematic representation of the SIMS process

5.3 Rutherford backscattering spectrometry (RBS)^{10,11}

The Rutherford backscattering spectrometry (RBS) technique relies in the elastic backscattering experienced by a light ion, typically He^+ , when colliding with the heavier atoms of the sample. The incoming ion loses an amount of energy proportional to the atomic mass of the atom it collides with. An energy detector, normally located at an angle of 170° , measures the energy of these backscattered ions (Figure II.8) [79]. This technique provides depth profiles of the sample. The signal detected from the backscattered ion is influenced by the energy loss dependent on the electron density and the distance travelled within the sample.

In this thesis, the evaluation of the RBS spectra was carried out using the SIMNRA software [80].

Figure II.8 Scheme of the RBS and ERDA analysis angles

¹⁰ The RBS measurements and analyses of the ScAlN thin films synthesized with the $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target have been carried out by the Accelerator Technique Group (ATG) of Evans Analytical Group (EAG).

¹¹ The RBS and ToF-ERDA measurements and analyses of the ScAlN thin films synthesized with the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target have been carried out in the “Groupe de recherche en physique et technologie des couches minces (GCM)”, University of Montréal; by Martin Chicoine.

5.4 Time of flight elastic recoil detection analysis (ToF-ERDA)

The elastic recoil detection analysis (ERDA) measures atoms that are sputtered from the sample surface by a heavy ion beam. This technique resembles RBS in that both rely in the elastic collisions between the incoming ion and the sample surface (I.2). However, whereas RBS uses light He^+ ions, this analysis technique measures the atoms that are sputtered from the sample surface by a high-energy (MeV) heavy-ion beam [81].

There are two complementary methods employed in ERDA for element identification. The mass of the detected atoms is identified by measuring both the energy and velocity of the recoiled atoms using time of flight (ToF) detectors. In this work, the evaluation of ToF-ERDA measurements has been carried out using the Allegria software [82].¹¹

6 Piezoelectric characterization

6.1 Piezoelectric force microscopy (PFM)

Similar in its operating principles to the atomic force microscopy (AFM), the piezoelectric force microscopy (PFM) is a scanning probe microscope (SPM) that detects voltage induced surface displacements. In this technique, a conductive tip probes the sample surface applying an electric field which induces local piezoelectric deformation [83]. Because of the converse piezoelectric effect, this deformation depends on the relative orientation between the polarization and the electric field vector.

This analysis provides information on the topography of the thin film surface, the amplitude of the piezoelectric response and its phase.¹²

6.2 Berlincourt piezometers¹²

The Berlincourt piezometers rely in the direct piezoelectric effect for measuring the piezoelectric d_{33} constant [84]. In this method, the sample is clamped and a low-frequency force is applied while the stress induced signal is recorded and compared to a reference sample (II.9). Note the measurement of the strain (S) and electric field (E) is performed at a constant stress (τ), which is necessary for measuring the piezoelectric d_{33} constant (II.4).

$$d_{33} = \left[\frac{\delta S_3}{\delta E_3} \right]_{\tau} \quad (\text{II.4})$$

¹² The piezoelectric force microscopy analyses were carried out using a JPK Nanowizard III AFM equipped with a PFM module and the piezoelectric constant d_{33} measurements were carried out using and a Berlincourt piezometer (PM300 Piezotest), by the “Emerging materials department, piezoelectric group” at Fraunhofer Institute for Applied Solid State Physics IAF.

Figure II.9 Scheme of a Berlincourt piezometer

7 Electrical characterization

The electrical characteristics of the fabricated SAW devices are measured using a vector network analyser (VNA)¹². The VNA measures the phase and amplitude of swept-frequency and swept-power test signals. In radio frequency (RF), because of the difficulties that arise when measuring the total current or voltage, the scattering parameters (S-parameters) are generally measured. These parameters resemble the reflection and transmission terms that are generally employed in optical systems.

S-parameters are of particular importance in the design of more complicated networks since the S-parameters of individual devices can easily be cascaded to predict the performance of more complex systems. The device admittance (Y) or impedance (Z) characteristics, usually measured in low-frequency device characterizations, can be derived from S-parameters.

7.1 Electrical properties of SAW devices

At resonance frequency the impedance of the motional branch is zero, meaning that most of the current goes through it. This resonance (f_s) is usually referred to in the literature as series or short-circuited resonance (II.5) for it requires the impedance to be zero.

$$f_s = \frac{1}{2\pi\sqrt{L_m C_m}} \quad (\text{II.5})$$

On the other hand, at the parallel (f_p) or antiresonance frequency (II.6), the impedance tends to infinite [17].

$$f_p = \frac{1}{2\pi\sqrt{\frac{L_m C_m C_0}{C_m + C_0}}} \quad (\text{II.6})$$

The effective acoustic velocity (v_{eff})

As it has been previously described, the acoustic velocity of a SAW depends on the density and elastic constants of the media through which it propagates. Thus, when the surface acoustic wave is generated in a heterostructure, the effective propagation velocity is set by a combination between the acoustic velocities of the different materials through which the wave travels. The effective velocity can be computed (II.7) using the series and parallel resonance frequencies [26][85].

$$v_{eff} = \lambda (f_p - f_s) / 2 \quad (II.7)$$

The effective electromechanical coupling coefficient (K_{eff}^2)

The effective electromechanical coupling coefficient measures the device efficiency in converting the mechanical energy into electrical energy and vice versa [17]. In SAW devices this parameter determines the separation between the resonance and anti-resonance frequencies (II.8).

$$K_{eff}^2 = \left(\frac{\pi^2}{2} \right) \left(\frac{f_p - f_s}{f_p} \right) \quad (II.8)$$

Unlike the electromechanical coupling coefficient of a piezoelectric material, this parameter measures the efficiency of the whole heterostructure i.e. substrate, IDT and piezoelectric layer. Using the parameters extracted from the mBVD model, the K_{eff}^2 can be computed using (II.9).

$$K_{eff}^2 = \left(\frac{\pi^2}{8} \right) \left(\frac{C_m}{C_0} \right) \left(\frac{C_0 - C_m}{C_0} \right) \quad (II.9)$$

The quality factor Q

By definition the quality factor Q is the ratio between the energy stored and the energy dissipated per cycle [17]. There are several methods proposed to calculate the Q factor from SAW resonators [86]. The -3 dB method (II.10) is widely employed because of its simplicity, however if the resonance and anti-resonance frequencies are close they “pull” each other, giving this formula overestimated values. By definition, this method is the ratio between the resonance frequency (series (s) or parallel (p)) and the frequency width at -3 dB.

$$Q_{s,p} = \frac{f_{s,p}}{\Delta f_{s,p-3dB}} \quad (II.10)$$

Other method that has been largely employed for SAW resonators is by analysing the steepness of the phase (φ) in the vicinity of the resonance frequencies (II.11) [87].

$$Q_{s,p} = \frac{f_{s,p}}{2} \left(\frac{\partial \varphi}{\partial f} \right)_{s,p} \quad (II.11)$$

On the other hand, the Q factor can be computed using the simulated parameters from the mBVD model (II.12):

$$Q_s = 2\pi f_s \frac{L_m}{R_m} \quad (II.12)$$

$$Q_p = \frac{1}{2\pi f_p C_0 R_0}$$

7.2 Modified Butterworth van Dyke (mBVD) model

The Butterworth van Dyke (BVD) model describes the electrical performance of SAW devices using the individual elements of an equivalent electrical circuit [17]. The BVD model does not account for the electrical losses presented in every SAW device. The modified Butterworth van Dyke (mBVD) model (II.9) includes additional dissipative elements R_s , C_0 and R_0 . The resistor R_s represents ohmic losses in the IDT. The substrate plate capacitance C_0 and the dielectric loss R_0 , respectively, describe the parasitic capacitance generated by the substrate and the dielectric losses in the heterostructure. All these elements will reflect on the S-parameters as higher reflection losses. Each resonance mode is simulated via a motional branch, which includes motional resistance, inductance and capacitance (R_m , L_m , C_m).

Figure II.9 mBVD model implemented in the Keysight advanced design system (ADS). It simulates three different resonance modes with three discrete motional branches

III ScAlN thin films: Results and discussion

In this chapter the studies carried out in the synthesis of ScAlN thin films are presented. The sputtering process in this thesis is carried out without intentional substrate heating, which further complicates the synthesis on substrates with a large surface roughness or when the lattice mismatch between the ScAlN compound and the substrate provokes the synthesis of highly stressed thin films.

The process described for preparing the sputtering system prior each deposition (see section II.2.4) ensured that the synthesis was carried out in a steady state regime. The steady state regime, monitored by the neglectable variations of the discharge voltage and discharge current, is achieved when the coverage of the alloy target surface is counterpart balanced by the sputtering process.

1 Compositional analyses: on the Sc shortage between the thin film and alloy target compositions

The different compositional techniques employed in this thesis report a reduction of the scandium content within the as synthesized $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films as compared to the alloy targets used (Figure III.1). This shortage of Sc has been previously reported in the synthesis of ScAlN thin from an alloy target in reactive sputtering [69]. Several of the analysis spectrums are shown below as an example of the measurements carried out.

Although the steady state in which the synthesis is carried out implies that the sputtering flux of the elements within the target is equal, it does not imply that the thin film composition to be equal to that of the target [75]. In the sputtering process, the distribution of material thorough the synthesis chamber depends on multiple process parameters such as the geometry of the synthesis chamber, the relative position between target and substrate, the angular distribution, sticking coefficient and atomic weight of the sputtered elements as well as the combination of process pressure and discharge process [74][88]. As it has been previously commented, the discharge power sets the energy of the sputtered atoms whereas the process pressure sets their mean free path between collisions and thus their scattering and thermalization during their travelling through the gas phase. The scattering of the sputtered atoms strongly depends on the atomic weight and radius. Light elements will lose their directionality in few collisions whereas large and heavy atoms do not deviate from its path after several collisions. Furthermore, due to the large scattering suffered by the lighter element, the alloy target is enriched with this element, slowly varying its composition [89]. All these have a significant influence when the reactive sputtering process is carried out from an alloy target.

During this thesis, the theoretical enrichment of the alloy target with the lighter element (Al) has not been reflected in a change of the thin film composition. Furthermore, the N_2 reactive gas reacts with the outermost atoms of the target and the composition of the alloy target surface varies. The geometry and configuration of the synthesis chamber set the position of the substrate on-axis with the alloy target. Altogether leads to the conclusion that the Sc content differences between the as synthesized thin films and the alloy target is due the angular distribution of the sputtered elements and their scattering during the gas phase.

Figure III.1 Composition of synthesized ScAlN thin films as measured from different analysis techniques.
 Left: $Sc_{0.26}Al_{0.74}N$ thin films synthesized with the $Sc_{0.40}Al_{0.60}$ alloy target. Right: $Sc_{0.43}Al_{0.57}$ thin films synthesized with the $Sc_{0.60}Al_{0.40}$ alloy target

The main drawback of measuring ScAlN thin films using the XEDS technique is that the representative $K\alpha$ line for N (0.3924 KeV) overlaps with the $L\alpha$ line for Sc (0.3955 KeV) and thus the $K\alpha$ line of Sc (at 4.0892 KeV) must be used for quantification. Figure III.2 shows the typical spectra of a XEDS analysis on a $Sc_{0.26}Al_{0.74}N$ thin films synthesized using the $Sc_{0.40}Al_{0.60}$.

Figure III.2 (Adapted from *Figure 3 of PAPER 1¹³*) XEDS analysis of an as-synthesized $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films on B-doped Si (001) substrate

The SIMS technique, as commented above, outstands for its sensitivity and because of the capability of detecting, with few exceptions, all the elements in the periodic table. Figure III.3 shows the SIMS analysis of a 2000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film.

Figure III.3 SIMS analysis of an as-synthesized $\text{Sc}_{0.41}\text{Al}_{0.59}\text{N}$ ScAlN thin film on a polycrystalline diamond substrate

RBS provides compositional and depth information of the elements within the sample. Figure III.4 shows the spectrum of a RBS analysis carried out in a $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a

¹³ Reprinted by permission from Springer Nature Customer Service Centre GmbH: Springer Nature, Microsystem Technologies, Synthesis of ScAlN thin films on Si (100) substrates at room temperature, A. Pérez-Campos, M. Sinusía Lozano, F. Javier García-García et al, 4661280082424, 2017

polycrystalline diamond substrate and the simulation carried out with the SIMNRA software [80]. Using this technique traces of heavy elements (not shown in Figure III.4) were detected. They were identified as Hafnium (Hf), a common contamination of rare-earth elemental targets [90][91].

Figure III.4 RBS analysis of a 2000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a polycrystalline diamond substrate

In this thesis, the ToF-ERDA technique has been employed because of its high sensitivity to light elements, which provided information about the oxygen content within the synthesized thin films. This analysis confirmed the presence of an oxide layer at the surface and that the oxygen concentration along the thin film is about 1% (Figure III.5).

Figure III.5 ToF-ERDA analysis of a 600 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a polycrystalline diamond substrate

2 Synthesis of highly c-axis oriented $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films

Employing the sputtering parameters shown in Table II.1 and Table II.2, ScAlN thin films were synthesized on B-doped Si (001) substrates. The texture of these thin films was analyzed using XRD θ -2 θ scans, whereas the FWHM of the rocking curve on the (0002) reflection was employed to study their degree of c-axis orientation.

Figure III.6 shows a typical θ /2 θ spectrum of a $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film synthesized on a polycrystalline diamond substrate. The absence of any other reflections but those corresponding to the ScAlN (0002) and (0004) indicates that the thin film is purely c-axis oriented.

Figure III.6 θ -2 θ analysis of a 100 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film on a polycrystalline diamond substrate

The discharge power provides the potential difference to ionize and accelerate the ions towards the target and therefore an increase of the discharge power rises the sputtering and deposition rates. The influence of the process pressure on the sputtering and deposition rate is related to the thermalization process experienced by the particles in the gas phase. Increasing the energy supplied to the growing thin film i.e. increasing the discharge power and reducing the process pressure, causes an improvement of the rocking curve FWHM value. However, the 900 W series of the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films do not follow this trend (Figure III.7 Left). A large discharge power is reported to degrade the quality of thin films because of the inherent increase of sputtering rate [32]. On the one hand, voids and defects are created within the growing thin film as the adatoms cannot diffuse towards their energetically favourable positions because their diffusion is blocked by a new layer of arriving species. On the other hand, these species reach the substrate with large kinetic energies which cause a knock-out effect degrading the microstructure quality of the thin film. Furthermore, depending on the synthesis conditions employed, highly compressive or highly tensile stressed thin films can be synthesized [92].

However, the degree of c-axis orientation of the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films is not improved at a combination of process pressure and discharge power of 0.33 Pa and 500 W and 0.40 Pa and 300 W (Figure III.7 Right). This is because the plasma is not stable in these synthesis conditions (arcing) interrupting the constant flow of species arriving on the thin film substrate.

Figure III.7 Influence of the process pressure and the discharge power on the FWHM of ScAlN thin films synthesized on B-doped Si (001) substrates using two different alloy targets

(Left: Adapted from PAPER 2¹⁴, FWHM values of the 900 W series obtained from PAPER 1¹⁵)

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¹⁵ Adapted by permission from Springer Nature Customer Service Centre GmbH: Springer Nature, Microsystem Technologies, Synthesis of ScAlN thin films on Si (100) substrates at room temperature, A. Pérez-

When dealing with the device fabrication process a rough thin film surface complicates the fabrication and compromises the electrical performance of the device. On the one hand, the adhesion of the metal employed in the IDT fabrication is reduced on rough surfaces. In regards to the SAW generation and propagation the most relevant loss mechanism for a SAW propagating through polycrystalline materials is scattering due to the inhomogeneities within the synthesized thin films [19]. The topography analyses revealed smooth surfaces with R_{RMS} values below 3 nm, whereas cross-section SEM images revealed a dense packed columnar growth of the thin films which agrees with the Zone T type in Thornton's structure-zone model (Figure III.8) [93].

Figure III.8 2 SEM cross section of a 2000 nm thick $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film synthesized on a polycrystalline diamond substrate. $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film synthesized with a process pressure of 0.40 Pa and a discharge power of 700 W

2.1 Influence of the gas admixture ratio in the synthesis of $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films

As it has been previously commented, a larger concentration of Ar within the gas admixture ratio increases the sputtering rate (Figure III.9). According to Lu *et al.* [94], the density of misoriented grains within the thin film varies with the N_2 concentration, and only when the synthesis is carried out in a pure N_2 atmosphere the density of these grains is neglectable. Therefore, in order to evaluate the influence of the gas admixture ratio on the texture and piezoelectric response, $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films were synthesized using the sputtering parameters shown in Table II.3. The combination of discharge power (500 W) and process pressure (0.40 Pa) were chosen after the results presented previously, as these synthesis parameters promote highly c-axis oriented $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.

Figure III.9 Deposition rate as a function of the gas admixture ratio. These syntheses were carried out at a discharge power of 500 W and a process pressure of 0.4 Pa using the $Sc_{0.60}Al_{0.40}$ alloy target

The as synthesized $Sc_{0.43}Al_{0.57}N$ thin films were analysed by means of θ - 2θ scans showing only (0002) and (0004) reflections. Afterwards the rocking curve FWHM value was studied to determine the influence that the different N_2 concentrations employed for the synthesis had on the degree of the thin film c-axis orientation (Figure III.10).

In the growing thin film, voids and defects are cured when the adatoms diffuse towards their most energetically favourable positions. However, when the synthesis is carried out at large deposition rates i.e. high Ar content within the gas admixture, the necessary diffusion process for curing the thin film is halted. This is observed as the rocking curve FWHM value increases from 3.08° to 4.23° when the argon content increases from 25 % to 75% in the 300 nm thick $Sc_{0.43}Al_{0.57}N$ thin films (Figure III.10 Right). However, as the thin film grows thicker i.e. more deposition time, the growing thin film cures these voids and defects and the role of the non-reactive gas content within the gas admixture ratio is dismissed. This is confirmed by the minimal differences of the rocking curve FWHM values ($2.18^\circ \pm 0.07^\circ$) obtained for the 1000 nm thick $Sc_{0.43}Al_{0.57}N$ thin films (Figure III.10 Left).

Figure III.10 Influence of the gas admixture ratio in the rocking curve FWHM value of 1000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrates

2.2 Evaluating the rocking curve FWHM values: a comparative analysis

As it has been previously commented, the syntheses of this work have been carried out without an intentional substrate heating, supplying the necessary energy to the adatoms for diffusing by means of the synthesis conditions, mainly by the combination of process pressure and discharge power. However, a comparison of these parameters is extremely difficult as they have to be optimized for each sputtering configuration and geometry to obtain highly c-axis oriented ScAlN thin films. In the literature, the process pressure and discharge power usually referred to range between 0.20-0.80 Pa and 100-1000 W respectively. Aiming to compare the measured rocking curve FWHM values obtained during this thesis with those in the literature, several parameters have been considered (Figure III.11). Firstly, due to the disparity in presenting the experimental results, only those studies specifying the substrate temperature, thin film composition and rocking curve FWHM measurements have been taken into account. In cases when the substrate temperature during the synthesis was reported as a temperature range (for example, 623-673 K), the figure only refers to the maximum temperature in the range (in the example: 673 K). Furthermore, the different substrates and seed layers employed are considered to have a smooth surface, since most are commercial substrates (Si (001) or Al_2O_3 (0001)), and metallic layers (either sputtered or evaporated) on thermally grown oxides (SiO_2). Furthermore, in order to present significant data, only the best rocking curve FWHM value per composition found in each study has been considered. Therefore, from this analysis several parameters are highlighted:

- Most of the syntheses in the literature are found to be carried out at substrate temperatures ranging from 350 °C to 550 °C.
- The median scandium content of the thin films in the literature is $\text{Sc}=22.5\%$.
- The co-sputtering configuration is the technique most widely employed.

Provided that the rocking curve FWHM values presented in this thesis are comparable with those in the literature, the added value of this synthesis conditions is that they are carried out with no intentional substrate heating. It implies cost and time savings during the synthesis, since heaters are not employed and the time between introducing the substrate into the sputtering system and beginning the synthesis is only constrained by the necessary vacuum condition within the load-lock chamber.

Figure III.11 Comparison between the rocking curve FWHM values and substrate temperature during the synthesis. Substrates: Si (*001*): This thesis, PAPER 1, PAPER 2, [8], [42], [43], [46], [69], [70], [94], [95], [96], [97]; Al₂O₃ (*0001*): [97], [98], [99]; Pt (*111*): [100], [101]; Mo (*110*): [44], [71], [100], [102]; TiN (*111*): [50], [90]; Ti (*0002*): [50]; AlCu (*111*): [50]; Polycrystalline diamond: This thesis, PAPER 3, PAPER 4, [103]. Broken parallel lines indicate overlapping of the adjacent points, which have been horizontally displaced

3 Piezoelectric analyses

3.1 $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films

The degree of c-axis orientation is not the only consideration when dealing with piezoelectric layers. Inversion domains within the thin film degrade their piezoelectric response. The polarity inversion in piezoelectric III-V thin films is reported to be promoted either by the introduction of O_2 into the gas admixture ratio or by increasing the discharge power [104][105][106]. In order to characterize completely the piezoelectric response of the thin film, PFM analyses were carried out (Figure III.12).

Within the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films inversion domains are found whereas the thin films are predominantly N-polar. The topography analyses on these thin films show grains protruding from the surface with no piezoelectric amplitude, reducing the overall piezoelectric response of the thin film (Figure III.12 above). The rocking curve FWHM value of this thin film is 3.05° .

On the other hand, the PFM analyses carried out on the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films (rocking curve FWHM value of 2°) show homogeneous N-polar thin films with no inversion domains. The mapping of the piezoelectric amplitude indicates a uniform piezoelectric response thorough the thin films (Figure III.12 below).

Figure III.12 (Above: Adapted from Figure 6 of PAPER 2¹⁷) 1x1 μm PFM analyses on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ (above) and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ (below) thin films synthesized on As-doped Si (001) substrates

3.2 $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films

The piezoelectric constant d_{33} of the samples employed to study the influence of the gas admixture ratio (see section III.2.1) was measured using a Berlincourt piezometer (Figure III.12). The

concentration of non-reactive gas within the gas admixture ratio has not a significant impact on the piezoelectric constant d_{33} . However, the 300 nm thin films present slightly lower values that vary between -16 pC/N and -18 pC/N whereas the 1000 nm $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films range from -19 pC/N and -24 pC/N. This is related to the influence of the thin film thickness on the d_{33} constant measurement; the 300 nm thick ScAlN thin films contain a lower amount of dipoles summing up for the piezoelectric response as compared to 1000 nm $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films.

The PFM analyses carried out on this set of samples agree with the Berlincourt piezometer measurements. Neither inversion domains nor misoriented grains degrading the piezoelectric response of the thin films were observed.

Figure III.13 Influence of the gas admixture ratio in the piezoelectric constant d_{33} of 1000 nm and 300 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrates

Furthermore, as it has been previously commented, the piezoelectric constant of the compound depends on its Sc concentration. This agrees with the maximum d_{33} constant measured using a Berlincourt piezometer, as the maximum measured value for the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films is -12 pC/N whereas the value corresponding to the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film duplicates it: -24 pC/N (Figure III.14).

Figure III.14 Maximum measured d_{33} constant of 1000 nm thick $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films synthesized on B-doped Si (001) substrate

4 Summary

In this chapter the influence on the piezoelectric response and texture of ScAlN thin films synthesized using the reactive sputtering technique with a $\text{Sc}_x\text{Al}_{1-x}$ alloy target is presented. The study carried out focuses mainly on the discharge power, process pressure and gas admixture ratio, synthesis parameters that are easily monitored and whose influence in the sputtering process had been widely reported for AlN thin films.

During this work, early experiments were carried out using the $\text{Sc}_{0.40}\text{Al}_{0.60}$ alloy target, with which $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films were synthesized and characterized. The Sc shortage within the thin film as compared to the alloy target is explained by the scattering of the sputtered elements in the gas phase and their angular distribution during sputtering. Afterwards the $\text{Sc}_{0.60}\text{Al}_{0.40}$ alloy target was purchased, and the indium employed to bond a 3 mm thick copper baking plate forced to not to use discharge powers above 500 W. With this alloy target, $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films were synthesized.

The discharge power provides, to a large extent, the kinetic energy to the ionized plasma species which impinge towards the target surface. In this collision their energy is transferred to the sputtered particles that travel through the gas phase towards the substrate surface. On the other hand, the process pressure sets the mean free path between collisions within the gas phase, which is the reason behind the thermalization of the sputtered particles. Both parameters play their role in the energy provided to the growing thin film, which is relevant since every synthesis in this thesis is carried out without an intentional substrate heating. However, to promote highly c-axis oriented ScAlN thin films the energy window provided during the synthesis by the overall sputtering parameters should be controlled, since a lack of or an excess of energy degrades the texture of the ScAlN thin film.

Regarding the gas admixture ratio, it has not a significant impact neither on the structural nor piezoelectric properties of the synthesized $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films. However, an increase of the thin film thickness improves the degree of c-axis orientation and the piezoelectric response of the thin films.

Therefore, when aiming the deposition of thin films with thicknesses below 300 nm, the synthesis should be carried out at discharge power and process pressures necessary to promote highly c-axis

oriented ScAlN thin films. On the other hand, the gas admixture ratio can be employed to tune the deposition rate.

Furthermore, the rocking curve FWHM values of the ScAlN thin films presented in this thesis are comparable to those found in the literature with an added value: the syntheses are carried out with no intentional substrate heating.

IV ScAlN based SAW devices

In this chapter the devices fabricated using the synthesized ScAlN thin films are presented. The $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films were synthesized on either thermally grown SiO_2/Si (001), a polycrystalline diamond layer synthesized by microwave plasma chemical vapour deposition (MPCVD) on top of 500 μm thick Si (001) or single crystal diamond (SCD) substrates, using the synthesis conditions shown in Table IV.1.

Table IV.1 Conditions employed for the synthesis of $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ (left) and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ (right) thin films.

$\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$		$\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$	
Discharge power (W)	700	Discharge power (W)	500
Process pressure (Pa)	0.53	Process pressure (Pa)	0.40
$\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)} (\%)$	25	$\frac{\text{Ar}}{(\text{Ar} + \text{N}_2)} (\%)$	25
Total gas flow $(\text{Ar} + \text{N}_2)$ (sccm)	12	Total gas flow $(\text{Ar} + \text{N}_2)$ (sccm)	12
Target-to-substrate distance (mm)	45	Target-to-substrate distance (mm)	45
Substrate temperature (K)	298	Substrate temperature (K)	298
Alloy target composition	$\text{Sc}_{0.40}\text{Al}_{0.60}$	Alloy target composition	$\text{Sc}_{0.60}\text{Al}_{0.40}$

Before carrying out the IDT fabrication process, the degree of c-axis orientation of the as synthesized ScAlN thin films was analyzed. Every ScAlN thin film, independently of the substrate employed, showed rocking curve FWHM values below 3° . Every device was inspected at each step of the fabrication process by SEM, whose nanoscale resolution allowed to discern failures. Every IDT presented in the following was fabricated with a metallization ratio of 0.5.

1 One port SAW resonators on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ based heterostructures

The $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ based SAW resonators were fabricated using an IDT made out of platinum (Pt) and a heterostructure comprising a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film and polycrystalline diamond substrates (Figure IV.1). According to the dispersion curves reported by Hashimoto *et al.* [25][27], the ratio between the piezoelectric thin film thickness and the IDT wavelength ($d/\lambda=2000/2800=0.71$) of these devices targets the maximum effective electromechanical coupling coefficient of the Sezawa mode. Furthermore, the device design targets the Sezawa mode resonance frequency at 2.70 GHz with a 2000 nm thick $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ nm within the polycrystalline diamond heterostructure.

Figure IV.1 Scheme of the SAW resonator fabricated using a Pt IDT on a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructure. IDT wavelength $\lambda=2800$ nm

1.1 Influence of the ScAlN thin film thickness

The acoustic velocity of a SAW depends on the density and elastic constants of the media through which the wave propagates. Thus, the predominant propagation of the generated SAW ($\lambda=2800$ nm), either through the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film or through the polycrystalline diamond substrate, sets the velocity at which the wave propagates within the heterostructure and therefore the frequencies of its resonance modes [19]. The shift of the Rayleigh and Sezawa modes of SAW resonators fabricated with three $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film thicknesses: 1700 nm, 2000 nm and 2300 nm is remarkable as observed in their S_{11} measurements (Figure IV.2).

The predominant capacitive behaviour of the fabricated SAW resonators as well as the impedance matching of the Sezawa mode with the characteristic impedance (50Ω) becomes apparent in the Smith chart representation (Figure IV.2 inset).

Figure IV.2 (Adapted from *Figure 4* of PAPER 4) Reflection coefficient of SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Superimposed: mBVD simulation result. Inset: Impedance Smith chart. d stands for the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film thickness

The fine fitting between the measured reflection coefficients and the mBVD simulation results validates the parameter extraction and their use for computing the effective SAW velocity, the effective electromechanical coupling and the quality factor of the devices (see PAPER 4).

The computed values agree with the trend of the dispersion curves reported by Hashimoto *et al.* [25][27]. However, as commented above, the structural and piezoelectric properties of the $\text{Sc}_x\text{Al}_{1-x}\text{N}$ compound are set by the scandium concentration (x) [39]. Thus, the slight mismatch between the mBVD simulation results and those reported in Hashimoto's simulation is caused by the different ScAlN thin film compositions (Hashimoto's simulations[25][27]: $\text{Sc}_{0.40}\text{Al}_{0.60}\text{N}$, these ScAlN thin films: $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$).

In regards to the effective SAW velocity, the Rayleigh mode approaches the characteristic velocity of the piezoelectric $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ compound (~ 4000 m/s) whereas the Sezawa mode velocity is largely affected by the increase of the ratio between the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film thickness and the designed IDT wavelength (Figure IV.3).

Figure IV.3 (Adapted from Figure 5 of PAPER 4) Effective SAW velocity of the Rayleigh and Sezawa modes generated in SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Lines are a guide for the reader

The Sezawa mode K_{eff}^2 coefficients experience a larger variation (23 %) than those of the Rayleigh mode (14%) for the devices fabricated using a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film thickness of 1700 nm, 2000 nm and 2300 nm (Figure IV.4). These results agree with the electromechanical coupling coefficient dispersion curves reported by Hashimoto *et al.* [25][27]. However, whereas the Rayleigh mode K_{eff}^2 coefficients agree in magnitude with those reported by Hashimoto *et al.*, the calculated Sezawa mode K_{eff}^2 coefficients are reduced by a 40 % as compared to those of the simulated dispersion curves.

Figure IV.4 (Adapted from Figure 6 of PAPER 4) Effective electromechanical coupling coefficient of the Rayleigh and Sezawa modes generated in SAW resonators fabricated on $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures with three different piezoelectric thin film thicknesses. Lines are a guide for the reader

1.2 Temperature coefficient of frequency (TCF)

The temperature coefficient of frequency (TCF) is the parameter that describes the frequency shift caused on a device by the temperature [19]. It must be considered in extreme working conditions such as in electronics for space applications where the devices are subjected to a wide range of temperatures [107]. This parameter is related to the temperature induced change of the elastic constants which modifies the acoustic velocity at which the wave propagates. Thus, combining materials with different thermal expansion coefficients, a zero-TCF device can be fabricated [108] [109].

In this study the SAW resonators fabricated in a 250 nm Pt/2000 nm $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructure, were electrically characterized in the 20 K- 450 K temperature range. The desorption of gas molecules adsorbed on the device surface influence their resonance frequency. For that reason the devices were characterized in a first temperature iteration from 20 K to 450 K and, after measuring the devices at the maximum temperature of the iteration i.e. 450 K, the temperature was sustained for 180 minutes before decreasing it to 20 K and characterize the SAW resonators in a second temperature iteration up to 300 K. The S_{11} measurements clearly show the frequency shift caused by the temperature on the Sezawa mode reflection coefficient (Figure IV.5).

Figure IV.5 (Adapted from PAPER 3¹⁶) Frequency shift of the Sezawa mode reflection coefficient due to the temperature variation of SAW resonators fabricated on 2000 nm $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures. Above: First temperature iteration; Below: Second temperature

The Sezawa mode effective acoustic velocities reflect the commented modification of the thin film elastic constant with the temperature (Figure IV.6). The differences between the effective SAW velocities of the first and the second temperature iterations are related to the desorption of gas (mainly water molecules) caused during the chuck's heating up process.

Figure IV.6 (Adapted from *Figure 5* of PAPER 3¹⁹) Temperature variation of the Sezawa mode effective acoustic velocities of a SAW resonator fabricated on 2000 nm $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures. Lines are a guide for the reader

The first order temperature coefficient of frequency (TCF) is defined as the variation of the frequency response of the resonator with the substrate temperature change (IV.1) [19][110].

$$TCF = \frac{1}{f_0} \frac{\partial f_0}{\partial T} \quad (\text{IV.1})$$

The TCF values range from 7.59 ppm/K at 20 K to -32.02 ppm/K at 450 K for the first temperature iteration showing a highly linear behaviour within two different temperature ranges, namely from 20 K to room temperature (300 K) as well as from 350 K up to 450 K (Figure IV.7). A similar linear trend can be appreciated in the TCF values for the second temperature iteration, where the calculated TCF values vary from 0 ppm/K at 20 K to -18.261 ppm/K at 300 K. These values report that the SAW resonators are stable in the measured temperature range and that they could be employed without using the thermal expansion compensation layer approach.

Figure IV.7 (Adapted from Figure 9 of PAPER 3¹⁹) Sezawa mode temperature coefficient of frequency of SAW resonators fabricated on 2000 nm $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ /polycrystalline diamond heterostructures. Lines are a guide for the reader

2 SAW devices on $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ heterostructures

The mass loading effect due to the finite IDT thickness varies the propagation velocities of the generated modes [111]. Platinum is usually employed in high temperature and harsh environments applications. It is considered a noble metal, thus chemically stable. Its oxidation-resistance derives from the protective oxidized thin film surface [112]. However, it is a heavy mass and highly resistive element as compared to other metal elements usually employed for the IDT fabrication. Among these gold and aluminum are largely employed. To reduce the resistivity of the IDT in this set of devices, a Cr adhesion layer and a gold thin film have been employed to fabricate the IDT.

2.1 One port SAW resonators

In this section, the electrical characterization of SAW resonators fabricated using an 110 nm thick gold IDT on a 2000 nm $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ piezoelectric layer are shown (Figure IV.8). A 5 nm chromium layer was used to improve the adhesion of the IDT to the piezoelectric thin film. In these designs the IDT wavelength was set to $\lambda=2800$ nm.

Three different substrates, namely thermally grown SiO_2 on Si (001), polycrystalline diamond/Si (001) and single crystal diamond (SCD), have been used to study their influence on the electrical characteristics of the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ based SAW resonators. SiO_2 thin films are usually employed as thermal expansion compensation layers in applications requiring temperature stability [108]. The high acoustic velocity of polycrystalline diamond makes this substrate a potential candidate for future 5G systems. On the other hand the single crystal diamond has been reported to reduce the insertion losses of SAW devices as there is less scattering due to the absence of grain boundaries [25][110].

The insertion loss of a SAW device is largely related to the IDT and the scattering of the propagating SAW [113]. This scattering occurs mainly in the grain boundaries and defects within the thin film [19][110]. Therefore, a rough substrate roughness degrades not only the electrical performance of the device but complicates the synthesis of highly c-axis oriented ScAlN thin films. The rocking curve measurements reporting values below 3° on each substrate disregarded the influence of the substrate roughness on the synthesis, however their roughness could reduce the

propagation characteristics of the SAW through the heterostructure and degrade the device electrical performance [110].

The thermally grown SiO_2 substrate is an electronic grade wafer and its surface is thus considered as atomic flat. The SCD datasheet report a R_a value below 2 nm. AFM analyses carried out on the polycrystalline diamond substrate report R_{RMS} values below 2 nm.

The devices were designed to work on a ratio between piezoelectric thin film thickness and IDT wavelength (d/λ) of 0.71. This ratio maximizes the K_{eff}^2 coefficient of the Sezawa mode for diamond based heterostructures [25][27]. Tang et al. [114] report dispersion curves for the ScAlN/Si stacking configuration which also shows a maximum of the Sezawa mode K_{eff}^2 coefficient around $d/\lambda=0.71$.

Figure IV.8 Schematic illustration of the SAW devices fabricated using three different $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ based heterostructures. IDT wavelength $\lambda=2800$ nm

$\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}/\text{SiO}_2$ heterostructure

Rayleigh (1.16 GHz) and Sezawa (1.80 GHz) resonance frequency modes are generated in the SiO_2 heterostructure (Figure IV.9). The reflection coefficient of both modes is below -20 dB indicating the great confinement of the SAW energy on the silicon based heterostructure. Silicon is disregarded in SAW applications due to the appearance of eddy currents [115]. This result is a very promising one regarding the integration of SAW devices in CMOS technology. It should also be noticed that, in the measured frequency range, the device insertion loss is extremely low as the baseline is close to 0 dB.

Figure IV.9 Reflection coefficient of a SAW resonator fabricated on the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}/\text{SiO}_2$ heterostructure

The capacitive behaviour of this resonator is shown in the Smith Chart in Figure IV.9 (Inset) as well as in the admittance characteristics shown in Figure IV.10.

Sc_{0.43}Al_{0.57}N/polycrystalline diamond heterostructure

The S_{11} characteristics of the Sc_{0.43}Al_{0.57}N/polycrystalline diamond heterostructure shows how several resonance frequency modes are generated together with second and third order reflections (Figure IV.11). Among these, the Rayleigh mode (1.20 GHz) reflection coefficient outstands (around -40 dB). The Sezawa mode (2.06 GHz) has a reflection coefficient of approximately -15 dB, whereas the second order Rayleigh mode (2.30 GHz) propagates with -18 dB at resonance. Furthermore, the insertion loss of this device is reduced as compared to those presented in the previous section with platinum IDTs.

Figure IV.11 Reflection coefficient of a SAW resonator fabricated on the Sc_{0.43}Al_{0.57}N/polycrystalline diamond heterostructure

The capacitive behaviour of this resonator is shown in the Smith Chart in Figure IV.11 (Inset) as well as in the admittance characteristics shown in Figure IV.12.

Figure IV.12 Admittance characteristics of a SAW resonator fabricated on the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /polycrystalline diamond heterostructure

$\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /single crystal diamond heterostructure

The devices with the lowest insertion losses are fabricated with the single crystal diamond based heterostructure. Furthermore, the reflection coefficient around -50 dB of the Sezawa mode (2.03 GHz) indicates the efficient excitation of this mode in the heterostructure (Figure IV.13). The Rayleigh mode (1.20 GHz) propagates with a reflection coefficient below -5 dB whereas its second order reflection mode (2.25 GHz) propagates with a reflection coefficient below -15 dB. Higher reflection modes above 2.50 GHz propagate in this heterostructure.

Figure IV.13 Reflection coefficient of a SAW resonator fabricated on the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /single crystal diamond heterostructure

The capacitive behaviour of this resonator is shown in the Smith Chart in Figure IV.13 (Inset) as well as in the admittance characteristics shown in Figure IV.14.

Figure IV.14 Admittance characteristics of a SAW resonator fabricated on the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /single crystal diamond heterostructure

Discussion

From the series and parallel resonance frequency modes of the admittance characteristics shown below (Figure IV.10, Figure IV.12, Figure IV.14) the effective acoustic velocity (II.7) and the effective K_{eff}^2 coefficients (II.8) have been computed (Table IV.2).

The displacement of the conductance baseline towards positive values indicates larger propagation losses [110]. Therefore, from the electrical measurements it can be concluded that the single crystal diamond based heterostructure has the lower propagation losses followed by the heterostructure comprising the SiO_2 substrate. The polycrystalline diamond based heterostructure shows a larger displacement of the conductance baseline due to the scattering on the grain boundaries. The acoustic velocities obtained in the three different heterostructures under study agree with the diamond and SiO_2 dispersion curves reported in the literature [25][27][114]. The use of polycrystalline diamond and single crystal diamond substrates increases by more than a 100 % the effective K_{eff}^2 coefficient, when compared with the values obtained for the SiO_2 heterostructure.

Table IV.2 Effective acoustic velocity and effective K_{eff}^2 coefficient obtained from the series and parallel resonances of the resonance frequency modes generated on the SAW devices with three different $Sc_{0.43}Al_{0.57}N$ heterostructures

Substrate	Mode	f_s [GHz]	f_p [GHz]	v_{eff} [m/s]	K_{eff}^2 [%]
SiO₂	Rayleigh	1.16	1.16	3279	1.15
	2nd	1.80	1.82	5093	1.73
Polycrystalline diamond	Rayleigh	1.20	1.22	3402	3.14
	Sezawa	2.06	2.09	5830	3.57
	2nd Rayleigh	2.27	2.30	6407	4.13
Single crystal diamond	Rayleigh	1.21	1.23	3425	3.39
	Sezawa	2.02	2.05	5725	3.64
	2nd Rayleigh	2.25	2.28	6361	2.98

Furthermore, the Sezawa mode effective K_{eff}^2 coefficients obtained in the polycrystalline diamond based heterostructure using a 2000 nm $Sc_{0.43}Al_{0.57}N$ thin film has been improved by a factor of 150% those obtained for the 2000 nm $Sc_{0.26}Al_{0.74}N$ thin film (Figure IV.15). This increment in the Rayleigh mode is of 650 %.

Figure IV.15 Effective electromechanical coupling coefficient of the Rayleigh and Sezawa modes obtained in the polycrystalline diamond heterostructures with 2000 nm thick $Sc_{0.26}Al_{0.74}N$ and $Sc_{0.43}Al_{0.57}N$ thin films

2.2 π type SAW filters

π type SAW filters (Figure IV.16) have been fabricated connecting three one-port resonators. One of these resonators was designed to support the series resonance f_s whereas the other two, were designed to generate the parallel resonance frequency f_p (see section II.7.1 for further details regarding these resonant modes).

Figure IV.16 Schematic of the π type SAW filters

The bandwidth of a SAW filter is determined by the K_{eff}^2 coefficient. In order to fabricate thin film based SAW filters that may cope with the bandwidth required in future 5G systems, and based on the results shown in Table IV.2, polycrystalline and single crystal diamond based heterostructures have been studied (Figure IV.17). The designed IDT wavelength and the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film thickness of these devices were set to $\lambda=1200$ nm and $d=850$ nm respectively ($d/\lambda=0.71$).

Figure IV.17 Schematic illustration of the SAW filters fabricated using $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ based heterostructures.
IDT wavelength $\lambda=1200$ nm

The transmission coefficient of the SAW filter fabricated using the diamond heterostructure presents the Rayleigh and Sezawa modes with their centre frequency (f_c) at 2.66 GHz and 4.88 GHz respectively (Figure IV.18). Both modes present a maximum gain above -6 dB whereas their cut-off frequencies are below -12 dB.

Figure IV.18 Transmission coefficient of a SAW filter fabricated on a $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ /polycrystalline diamond heterostructure

On the other hand, the band pass maximum of the Rayleigh mode in the SAW filter fabricated on the single crystal diamond heterostructure is -2.5 dB (Figure IV.19). The centre frequency of this mode is 2.17 GHz whereas the Sezawa mode f_c is at 4.88 GHz and its band pass a maximum is -5 dB. The cut-off of both lower (f_1) and higher (f_2) frequencies in the Rayleigh mode is below -18 dB whereas those corresponding to the Sezawa mode are below -10 dB.

Figure IV.19 Transmission coefficient of a SAW filter fabricated on a $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film/single crystal diamond heterostructure

The -3 dB bandwidth of the Rayleigh mode is larger in the polycrystalline diamond heterostructure (Table IV.3). However, the Sezawa mode presents a larger bandwidth and centre frequency in the SAW filter fabricated using the single crystal diamond. The differences between the centre frequencies are due to the mass loading effect because the higher thickness of the IDT in the single crystal diamond heterostructure.

Table IV.3 -3 dB bandwidth and centre of frequency f_c of the Rayleigh and Sezawa modes in SAW filters fabricated on the two different $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ heterostructures

Heterostructure substrate	Mode	f_1 [GHz]	f_2 [GHz]	f_c [GHz]	-3 dB Bandwidth [MHz]
Polycrystalline diamond	Rayleigh	2.61	2.72	2.66	107
	Sezawa	4.64	4.83	4.74	189
Single crystal diamond	Rayleigh	2.14	2.21	2.17	72
	Sezawa	4.81	4.99	4.90	181

3 Summary

SAW resonators and filters have been fabricated using $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films on different heterostructures.

Using platinum IDTs the feasibility of using the ScAlN compound in SAW applications within the GHz range was studied. The proposed heterostructure comprises a substrate with high propagation

velocity (polycrystalline diamond), a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film and the aforementioned IDT made out of platinum. Furthermore, the temperature characteristics of the heterostructure were characterized in the 20 K–450 K temperature range, reporting that this heterostructure can be employed in applications with large temperature variations.

Due to the relationship between the heterostructure K_{eff}^2 coefficient and the d_{33} constant of the piezoelectric thin film, heterostructures comprising $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films were fabricated. These heterostructures comprised a 2000 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film with the IDT made out of a 5 nm Cr as adhesion layer and 110 nm of gold. The different substrates employed: a thermally grown SiO_2 , a polycrystalline diamond and a single crystal diamond, influence the electrical response of the device. Due to the absence of scattering in the grain boundaries, the insertion losses are almost neglectable for the device fabricated on the single crystal diamond whereas the largest losses are obtained for the polycrystalline diamond heterostructure.

Furthermore, the Sezawa mode K_{eff}^2 coefficient increases by a 125 % whereas the Rayleigh mode K_{eff}^2 improves in a 650 % in the polycrystalline diamond based heterostructure when the thin film composition is varied from $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ to $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$.

The Rayleigh and Sezawa modes in the SiO_2 based heterostructure show reflection coefficients around -20 dB comparable to the reflection modes generated in the other two heterostructures. However, the heterostructures comprising the polycrystalline and single diamond substrates improve the acoustic velocity and K_{eff}^2 coefficients obtained for the SiO_2 based heterostructure with little differences on these values between them.

For that reason, SAW filters were fabricated on the polycrystalline and single diamond based heterostructures. Both show larger bandwidths for the Sezawa than for the Rayleigh modes. This agrees with the theoretical (dispersion curves) and experimental results presented above, as the Sezawa K_{eff}^2 coefficients are larger than those of the Rayleigh mode in these heterostructures.

V Conclusion

The main achievements and conclusions within the work carried out in this thesis are described in this chapter. These results are presented according to how this thesis was divided.

Reactive sputtering of ScAlN thin films

A reproducible method to synthesize highly c-axis oriented $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ and $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films has been developed. The process pressure and the discharge power emerge as the sputtering parameters that influence the most in providing the energy to the adatoms for promoting the synthesis of the compound wurtzite phase. The PFM analyses report that discharge powers above 700 W facilitates the appearance of inversion domains which degrade the piezoelectric response of the thin film.

Independently of the substrate employed, the rocking curve FWHM values below 3° of the as synthesized ScAlN thin films and the columnar structure observed, reflects that the ScAlN compound can be synthesized with no intentional substrate heating with high crystallinity quality. Furthermore, the values presented in this thesis are comparable to rocking curve FWHM values of ScAlN thin films synthesized at substrate temperatures above 300°C .

The gas admixture ratio does not influence neither the texture quality nor the piezoelectric response of ScAlN thin films above 1000 nm. However, it can be a parameter to monitor the deposition rate and texture of thinner ScAlN thin films.

Furthermore, the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ d_{33} constant (-24 pC/N) duplicates the value obtained for the $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin films. This confirms the reported increase of the compound piezoelectric d_{33} constant with the thin film Sc content.

Design and fabrication of SAW devices

The feasibility of using ScAlN thin films in electroacoustic applications has been demonstrated fabricating SAW devices on different heterostructures. The large reflection coefficients in the generated Rayleigh and Sezawa modes together with the effective K_{eff}^2 coefficients obtained confirms it.

The stability of the SAW resonators in cryogenic temperatures has been studied obtaining the temperature coefficient of frequency. That particular heterostructure, which consist of a platinum layer for the IDT on a $\text{Sc}_{0.26}\text{Al}_{0.74}\text{N}$ thin film synthesized on a polycrystalline diamond substrate compensates the expansion coefficients of the different materials of the stacking configuration, providing high thermal stability up to 450 K, in which the electrical response of the device begins to degrade.

Chapter V

The insertion losses of the SAW resonators, which are largely bound to the IDT metal, have been reduced using gold instead of the more resistive platinum. Furthermore, the application of several different heterostructures comprising different substrates has demonstrated that the scattering due to the grain boundaries in the substrate is a parameter to consider in the design of SAW devices. Among the different substrates employed, the single crystal diamond outstands as the insertion losses obtained in its heterostructure are neglectable. Furthermore, Rayleigh and Sezawa mode effective K_{eff}^2 coefficients are above 3 % in the heterostructures with polycrystalline and single crystal diamond.

With the technology developed to fabricate and characterize the SAW resonators, bandpass filters have been fabricated with bandwidths of the Sezawa mode above 100 MHz in frequencies around 5 GHz, which definitely confirms the main objective of this thesis: *the feasibility of the ScAlN compound for being used in SAW devices for 5G applications.*

VI Future work

The work carried out in this thesis suggests several different research trends in order to gain further knowledge on the mechanisms in which the SAW is generated and propagated on the ScAlN compound, the performance and design of the devices and their future applications. Several ideas that have been planned or are being already carried out are presented in the following:

ScAlN compound

- Analysis of the first stages of the thin film synthesis by transmission electron microscopy (TEM). For this technique, the thickness of the specimen is usually a thin film below 100 nm, which would provide atomistic resolution on how the ScAlN compound begins its growth on different substrates and the mechanisms of energy release carried out as the thin film grows [100][116].
- Longitudinal and shear excitation of the SAW depend on the material through it propagates. There have been several studies reporting a directionality due to the c-axis tilt in ScAlN thin films [117][118]. Furthermore, giant effective K_{eff}^2 coefficients have been reported with a 33° tilted c-axis ScAlN compound [119]. The tilt of the c-axis should be studied by XRD and the synthesis parameters monitored to find a method to control the synthesis with a preferred inclination [120].

SAW devices

- Without modifying the designed IDT wavelength, the device resonance frequency can be tuned by the ScAlN thin film thickness, as the acoustic velocity of the wave propagating through the IDT/ScAlN/diamond heterostructure increases when a larger fraction of the wave travels through the faster diamond layer. Some work has been carried out in this regard and SAW filters with polycrystalline diamond based heterostructures comprising 200 nm thick $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin films have been fabricated. Their Rayleigh and Sezawa modes have a centre frequency around 4.0 and 7.5 GHz respectively (Figure VI.1). However, a deeper knowledge of the first stages of the synthesis is still needed to further improve the device performance.

Figure VI.1 Transmission coefficient of a SAW filter fabricated with IDT of 65 nm Au and 5 nm of Cr as adhesion layer on a 200 nm $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film/polycrystalline diamond heterostructure

- According to the theoretical work of Zhang *et al.* [26], embedding the IDT significantly increases the effective K_{eff}^2 coefficients of the heterostructure (Figure VI.2 Left). However, several technological problems must be addressed. First, the contact between the growing piezoelectric thin film and the IDT must be intimate. If not, voids and defects are created in the interface and the SAW is not efficiently generated. Second, if the piezoelectric thin film grows highly stressed, the IDTs can be peeled off. This stress can be generated by the synthesis conditions or the lattice mismatch between substrate and IDT materials with the ScAlN compound. SEM inspection of the device must be carried out to ensure the complete coverage of the IDT with the $\text{Sc}_{0.43}\text{Al}_{0.57}\text{N}$ thin film (Figure VI.2 Centre and right).

Applications

- SAW devices are widely used in sensor applications due to their high stability in harsh environments and measurement resolution. The devices presented in this work can be integrated as pressure or temperature sensors with a proper calibration. Furthermore, the SAW devices can be employed in lab-on-a-chip applications with nanomolar resolution [4].
- The increase of the electromechanical coupling coefficient using the ScAlN compound instead of AlN can be employed to fabricate plasmonic devices. In these devices the SAW facilitates the coupling of light and electrons leading to surface plasmon polaritons (SPPs) in 2D materials as graphene or the hexagonal boron nitride (h-BN) [6]. The efficiency of these devices is directly related to the amplitude of the propagating SAW. Furthermore, the propagation characteristics of the heterostructures presented in this work, denote that they can be the foundation to integrate plasmonic devices in Si based technologies.
- The strain field created by a propagating SAW has been reported to allow the modulation of phonons. Irikawa *et al.* reported this modulation using ZnO piezoelectric thin films whose properties are largely surpass by the ScAlN compound [121]. The heterostructures presented in this work present low insertion losses and high reflection coefficients which can be employed in these promising devices [122][123].
- Spintronics have attracted the attention of the EU council and they are funding several different projects which are reporting promising results in the field [124][125]. This technology couple the SAW to the spin which strengthen the spin current and reduces the current necessary to rotate the spin [7]. Integrating the magnetic nanowire in the path of the propagating SAW with the heterostructures presented in this work would allow the integration of spintronic based devices in CMOS technology.

VII References

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