Introduction

Transition metal nitride thin film are widely investigated for their mechanical [1–4], plasmonic [5], piezoelectric properties [6–8], or used as diffusion barriers [9, 10] and more recently for their thermoelectric properties [11]. For physical vapor deposition (PVD) such as sputtering or arc deposition, it is important to control the purity of the final film. With such techniques, high vacuum or ultra-high vacuum chambers with base pressure of $10^{-6}$ to $10^{-8}$ Pa are used to minimize the contaminations during the deposition process. Different aspects of the deposition process can be controlled in order to minimize contamination, such as the cleaning processes of chamber and substrates, bake-out, base pressure and purity of target/gas. Nevertheless, even at the best conditions of cleanliness, some transition metals which are more sensitive to others to contamination (oxygen, carbon) will lead to incorporation of impurities in the film during deposition.

Abstract

ScN is an emerging semiconductor with an indirect bandgap. It has attracted attention for its thermoelectric properties, use as seed layers, and for alloys for piezoelectric application. ScN and other transition metal nitride semiconductors used for their interesting electrical properties are sensitive to contaminants, such as oxygen or fluorine. In this present article, the influence of depositions conditions on the amount of oxygen contaminants incorporated in ScN films were investigated and their effects on the electrical properties (electrical resistivity and Seebeck coefficient) were studied. Epitaxial-like films of thickness $125 \pm 5$ nm to $155 \pm 5$ nm were deposited by DC-magnetron sputtering on c-plane Al$_2$O$_3$, MgO(1 1 1) and r-plane Al$_2$O$_3$ at substrate temperatures ranging from 700 $^\circ$C to 950 $^\circ$C. The amount of oxygen contaminant in the film, dissolved into ScN or as an oxide, was related to the adatom mobility during growth, which is affected by the deposition temperature and the presence of twin domain growth. The lowest values of electrical resistivity of $50 \, \mu\Omega \cdot \text{cm}$ were obtained on ScN(1 1 1)/MgO(1 1 1) and on ScN(0 0 1)/r-plane Al$_2$O$_3$ grown at 950 $^\circ$C with no twin domains and the lowest amount of oxygen contaminant. At the best, the films exhibited an electrical resistivity of $50 \, \mu\Omega \cdot \text{cm}$ with Seebeck coefficient values maintained at $-40 \, \mu\text{V} \, \text{K}^{-1}$, thus a power factor estimated at $3.2 \times 10^{-3} \, \text{W m}^{-1} \, \text{K}^{-2}$ (at room temperature).

Keywords: ScN, thin films, sputtering, x-ray spectroscopy, thermoelectricity

(Some figures may appear in colour only in the online journal)
Among the transition metals, scandium has one of the highest affinities for oxygen [12]. The presence of oxygen in the final film occurs most likely because of incorporation during the deposition process [12], rather than as a result of oxidation of the film by grain boundary diffusion during post-growth exposure to air [13]. Another source of contaminants for ScN is the target. Due to the production process, scandium often contains fluorine impurities which are typically found in ScN films [14–18]. Oxygen impurities incorporated into ScN influence the electronic properties of ScN films [12, 14, 16, 19–24]. Scandium nitride, as many of the transition metal nitride materials, has a rock-salt structure (B1) and exhibit high hardness and high melting point [25, 26]. ScN is a degenerate semiconductor with an indirect bandgap of 0.9–1.6 eV [12, 22] and shows n-type behavior with carrier concentration varying from 10^{18} to 10^{20} cm^{-3} [22, 27]. The presence of defects, impurities or variation of composition into ScN may affect drastically the electronic and thermoelectric properties of the final film.

For thermoelectrics, control of contaminants is essential for control of the electrical properties of the film. The efficiency of a thermoelectric material is defined by its dimensionless figure of merit, ZT = \frac{\sigma T}{\kappa} - \frac{1}{T}, where S is the Seebeck coefficient, \sigma is the electrical conductivity, \kappa is the thermal conductivity and T is the absolute temperature [28, 29]. The higher the thermoelectric figure of merit, i.e. the higher the power factor (S^2\sigma) and/or the lower the thermal conductivity, the more efficient the energy conversion. The origin of the large variation of thermoelectric properties obtained in the literature on ScN can be explained by a modification of the density of states (DOS) of ScN around the Fermi level with respect to the presence of points defects or impurities [30]. Kerdsongpanya et al have shown theoretically the possibility to alter the thermoelectric properties by the presence of contaminants [30]. Incorporation of oxygen (≤2 at.%) into the ScN cell causes a shift of the Fermi level into the conduction band without affecting the DOS enhancing the carrier concentration and electrical conductivity [11, 14, 20, 21, 30]. When the amount of contaminants increases, these electrical properties may deteriorate, and at even higher level, secondary phases such as Sc_2O_3-x, are formed [12, 14, 16, 20, 31, 32]. Our previous study on wet-cleaning of MgO(001) substrates have shown the important influence of the substrate cleaning process used prior to deposition on the reduce of scandium oxide formation into the film [31]. Generally, for ScN deposited by reactive sputtering, the lower the base pressure of the chamber, the lower is the content of oxygen incorporated into ScN [12, 20, 21]. Several papers [12, 14, 16, 20, 21, 25, 30–33] reported oxygen contaminations from below one to several percent into ScN but there is still a need to elucidate the effect of oxygen incorporation on the growth mechanism and the effect of the orientation of the film and the presence or not of twin domains during growth. Understanding of the oxygen incorporation into ScN film is a key point for control and improvement of its electrical and thermoelectric properties.

In this study, ScN thin film were studied to improve the understanding of the film growth process and the control of incorporation of oxygen. Different conditions of temperature (700 °C to 950 °C) and different substrates (c-plane Al_2O_3, MgO(111) and r-plane Al_2O_3) were used to deposit ScN film in a high vacuum chamber. Influence of the deposition conditions as well as the substrates used influences the incorporation of oxygen into ScN cell and/or oxide. The substrates used for this study were chosen for their dielectric character allowing further electrical and thermoelectric characterization, and because they are also commonly used to deposit thermoelectric thin films. Different orientations were considered in order to grow the film along different orientations and deposit the films under different growth mode. The effect of the temperature used, surface terminated atoms, twin domain growth, adatom mobility during deposition and oxygen content on the electrical and thermoelectric properties of ScN is investigated.

Materials and methods

ScN thin films were deposited using DC reactive magnetron sputtering in a high vacuum chamber (10^{-7} Pa) using a 2-inch Sc target (MaTek: Sc 99.5%) in an Ar/N_2 (flow ratio 75% Ar/25% N_2) sputtering-gas mixture. The pressure during depositions was kept at 0.27 Pa (2 mTorr) as well as the power at 125 W. The chamber is described elsewhere [34]. 10 mm × 10 mm one side-polished substrates of c-plane Al_2O_3, MgO(111) and r-plane Al_2O_3 (Alineason Materials & Technology) were used and simultaneously inserted in the chamber, on the same holder for deposition. The films were deposited during 60 min under constant rotation and at three different temperatures of deposition (T_D): 700, 820 and 950 °C. Prior to deposition the substrates were cleaned using detergent steps, then 10 min with acetone in ultrasonic bath and then repeated with ethanol and blown dry with a N_2-gun. The detergent steps are described elsewhere [31].

Structural characterization was performed by x-ray diffraction (XRD) on a PANalytical X’Pert PRO in Bragg Brentano configuration (with continuous rotation of the sample during the measurement) equipped with a Cu Kα radiation source and a four-circle texture instrument (PANalytical X’Pert MRD) equipped with a parallel beam Cu Kα1 radiation in θ − 2θ, ω and φ scan modes and for the x-ray reflectivity measurement. The morphology and the cross sections of the film were observed by a scanning electron microscope (SEM, LEO Gemini 1550, Zeiss).

XPS spectra were obtained using an Axis Ultra DLD instrument from Kratos Analytical (UK) with the base pressure during spectra acquisition of 1.1 × 10^{-7} Torr (1.5 × 10^{-7} Pa), with a monochromatic Al Kα radiation (hν = 1486.6 eV). The anode power was set to 150 W. Prior to analyses all samples were sputter-cleaned with 0.5 keV Ar+ ion beam incident at the 20° angle from the surface and rastered over the area of 3 × 3 mm^2. All spectra were collected from the area of 0.3 × 0.7 mm^2 and at normal emission angle using a low-energy electron gun to compensate for the sample charging. The analyzer pass energy was set to 20 eV which results in the full width at half maximum of 0.55 eV for the Ag 3d_{5/2} peak.
XPS data were treated using KoI XD fitting software [35]. To avoid doubts related to using the C 1s peak of adventitious carbon as a charge [36, 37], all spectra were aligned to the N 1s peak of Sc-N set at 396.5 eV. The latter procedure results in N 1s and Sc 2p binding energy values which are consistent with the NIST data base [38]. Peak fitting was performed for all spectra using Voigt functions and a Shirley background. Details of the peak fitting procedure are described in the supplementary information (available online at stacks.iop.org/JPhysD/52/035302/mmedia).

The electrical resistivity of the sample was determined indirectly by measuring the sheet resistance of the film with a four-point probe Jandel RM3000 station which was multiplied by the thickness of the films obtained from the cross-sectional SEM imaging. The Seebeck voltage was measured on a homemade Seebeck voltage measurement setup and performed at atmospheric pressure. The samples were electrically isolated on which two copper electrodes separated by 8 mm were connected to the surface of the sample (film) and connected to a multimeter with a resolution of 0.01 mV. A temperature measurement was performed to the surface of the sample (film) and connected to a multimeter with a resolution of 0.01 mV.

The mass density deduced from the x-ray reflectivity measurements varies from 3.75(1) g cm⁻³ at low temperature to 4.09(1) g cm⁻³ at high temperature. The densities obtained can be compared to the ScN bulk values of 4.25(2) [39]. The film deposited on c-plane Al₂O₃ does not exhibit a clear difference of the cell parameter when the TD increases from 700 °C to 950 °C with a value close to 4.504(4) Å. The ScN cell parameter deposited on MgO(1 1 1) increases from 4.503(4) Å at TD of 700 °C to 4.512(4) Å at 850 °C and stay constant for higher temperature. A similar observation can be made for the film deposited on r-plane Al₂O₃. The crystal quality can be evaluated from the measurement of the Δω and Δϕ on the ω-scans and φ-scans of the preferential orientation. The lower the values of the Δω and Δϕ, the higher the crystalline quality for a certain orientation. All films exhibited diffraction peaks on the φ-scans demonstrating a degree of ordering of the grains in the plane of the substrate. The term ‘epitaxial-like’ can be used to describe this, meaning that all grains are epitaxially related to the substrate, and domain-growth with two different stacking sequences occurs, but there is no global epitaxy. The crystal quality differs between substrates, with a higher crystal quality on c-plane Al₂O₃ and r-plane Al₂O₃ substrates than on MgO(1 1 1) substrate. Nevertheless, all films are epitaxially-like grown with higher quality at higher TD observed by a reduction of the Δω and Δϕ.

The detailed results from the XRD analysis and the thickness values are listed in table 1. The thickness is reduced from 155 ± 5 nm at a TD of 700 °C to 125 ± 5 nm at a TD of 950 °C. The mass density deduced from the x-ray reflectivity measurements varies from 3.75(1) g cm⁻³ at low temperature to 4.09(1) g cm⁻³ at high temperature. The densities obtained can be compared to the ScN bulk values of 4.25(2) [39]. The film deposited on c-plane Al₂O₃ does not exhibit a clear difference of the cell parameter when the TD increases from 700 °C to 950 °C with a value close to 4.504(4) Å. The ScN cell parameter deposited on MgO(1 1 1) increases from 4.503(4) Å at TD of 700 °C to 4.512(4) Å at 850 °C and stay constant for higher temperature. A similar observation can be made for the film deposited on r-plane Al₂O₃. The crystal quality can be evaluated from the measurement of the Δω and Δϕ on the ω-scans and φ-scans of the preferential orientation. The lower the values of the Δω and Δϕ, the higher the crystalline quality for a certain orientation. All films exhibited diffraction peaks on the φ-scans demonstrating a degree of ordering of the grains in the plane of the substrate. The term ‘epitaxial-like’ can be used to describe this, meaning that all grains are epitaxially related to the substrate, and domain-growth with two different stacking sequences occurs, but there is no global epitaxy. The crystal quality differs between substrates, with a higher crystal quality on c-plane Al₂O₃ and r-plane Al₂O₃ substrates than on MgO(1 1 1) substrate. Nevertheless, all films are epitaxially-like grown with higher quality at higher TD observed by a reduction of the Δω and Δϕ.

Figure 2 shows SEM images of the film surface morphologies. The surface morphology of the film grown on the three substrates at 950 °C is dense and smooth. The films on c-plane Al₂O₃ and MgO(1 1 1) are composed of small pyramidal shaped grains of 10–15 nm characteristic of a (1 1 1) oriented cubic symmetry material. On c-plane Al₂O₃, the dark areas between grains observed at the surface of the film are due to the twin domain growth of the film as opposed to the one on MgO(1 1 1) substrate. The film deposited on r-plane Al₂O₃ is composed of regular well-organized square-shaped grains with a size of 35–40 nm, characteristic of a (0 0 1) oriented cubic symmetry material. The morphology of the films deposited at 750 °C present a rough surface with large grain size. For all three substrates, the crystallites present a certain disorder at the surface of the film which shows a trend of texturing along the [1 1 1] or [0 0 1] directions. A film deposited at a TD of 820 °C can be described as an intermediate situation.

Figure 3 displays the detailed XPS spectra of the Sc 2p and N 1s core levels together with the deconvoluted peaks. The N 1s peak is composed for all films of a main N-Sc peak at 396.5 eV and a shoulder peak on the high-BE side (397.4 eV) corresponding to nitrogen in an oxynitride (N-(ScO,N₁₋₃)). The Sc 2p core level peak has several contributions: ScN peak characteristic of the ScN material situated at 400.8 eV [38]; a Sc-N peak of substoichiometric ScₙNₓ (x < 1) at 399.8 eV formed as a result of preferential sputtering of lighter N atoms.
The oxygen present in the film either as ScO$_x$ or dissolved in ScN (ScO$_x$N$_{1-x}$) gives rise to two more peaks in the Sc 2p spectra at 403.3 and 402.2–402.9 eV, respectively. With increasing growth temperature, the ScN$_x$ peak becomes more intense as the effective area cleaned by the Ar$^+$ beam increased due to the reduced surface roughness. This increase of temperature is also responsible for a decreasing intensity of the Sc 2p components of the oxide and oxynitride.

Figure 4 displays the detailed O 1s XPS spectra together with the peak models. Clearly, numerous contributions indicate that oxygen is present in several chemical states. A peak at 530.4 eV is assigned to the oxide ScO$_x$, followed by a higher BE component at 532.2 eV due to oxynitride ScO$_x$N$_{1-x}$. Finally, the peak at the highest BE (533.8 eV) is due to the contaminating species including CO, CO$_2$, and H$_2$O which remain at the surface due to incomplete cleaning of rougher films, as described above. The later component disappears in case of the 950$^\circ$C sample, which has the lowest surface roughness, and, hence, can be properly cleaned with the Ar$^+$ ion beam.

Table 1. Details from XRD analysis of ScN films deposited at different temperatures (700$^\circ$C, 820$^\circ$C and 950$^\circ$C) on: c-plane Al$_2$O$_3$, MgO(1 1 1) and r-plane Al$_2$O$_3$. Preferential orientations of the film; cell parameter calculated from the main peak; associated $\omega$-scan and $\varphi$-scan FWHM values of the preferential orientation and thickness are reported.

| Deposition temperature ($T_D$) | Substrate | ScN preferential orientation | $a_{ScN}$ (Å) | $\omega$-scan FWHM ($\Delta\omega$ in $^\circ$) | $\varphi$-scan FWHM ($\Delta\varphi$ in $^\circ$) | XRR density (g cm$^{-3}$) | Thickness (nm) |
|-------------------------------|-----------|-----------------------------|---------------|---------------------------------|--------------------------|------------------|----------------|
| 950$^\circ$C                   | c-plane Al$_2$O$_3$ (1 1 1) | 4.503          | 0.4  | 1.6 | 4.09 | 120 |
| 820$^\circ$C                   | MgO(1 1 1) (1 1 1) | 4.512          | 1.5  | 2.9 | 4.08 | 130 |
| 700$^\circ$C                   | r-plane Al$_2$O$_3$ (0 0 1) | 4.509          | 1.4  | 1.7 | 4.09 | 125 |
| 950$^\circ$C                   | c-plane Al$_2$O$_3$ (1 1 1) | 4.504          | 0.8  | 2.6 | 3.91 | 130 |
| 820$^\circ$C                   | MgO(1 1 1) (1 1 1) | 4.512          | 1.6  | 3.3 | 4.01 | 120 |
| 700$^\circ$C                   | r-plane Al$_2$O$_3$ (0 0 1) | 4.509          | 1.3  | 1.7 | 4.01 | 140 |
| 950$^\circ$C                   | c-plane Al$_2$O$_3$ (1 1 1) | 4.503          | 1.0  | 3.6 | 3.75 | 160 |
| 820$^\circ$C                   | MgO(1 1 1) (1 1 1) | 4.503          | 2.1  | 5.1 | 3.85 | 150 |
| 700$^\circ$C                   | r-plane Al$_2$O$_3$ (0 0 1) | 4.505          | 2.1  | 3.4 | 3.81 | 155 |

The oxygen present in the film either as ScO$_x$ or dissolved in ScN (ScO$_x$N$_{1-x}$) gives rise to two more peaks in the Sc 2p spectra at 403.3 and 402.2–402.9 eV, respectively. With increasing growth temperature, the ScN$_x$ peak becomes more intense as the effective area cleaned by the Ar$^+$ beam increased due to the reduced surface roughness. This increase of temperature is also responsible for a decreasing intensity of the Sc 2p components of the oxide and oxynitride.

Figure 5 displays the summary of the information extracted from the XPS analysis performed on the nine ScN films (see figures 3 and 4). Figure 5(a) displays the total fraction of scandium in an oxynitride ScO$_x$N$_{1-x}$ (x > 0) plus oxide ScO$_x$ and figure 5(b) represents the ScN$_x$ fraction detected in the Sc 2p or O 1s peak. The films on r-plane Al$_2$O$_3$ and on MgO(1 1 1) grown at 950$^\circ$C contain the lowest amount of Sc bonded to oxygen (only 4% of the total area under the Sc 2p spectra), of
which 95% corresponds to an oxynitride $\text{ScO}_{x-\delta}$ ($x > 0$).

For the film on $c$-plane $\text{Al}_2\text{O}_3$ grown at 950 °C, 30% of Sc atoms is bonded to oxygen, of which 60% are in the oxynitride $\text{ScO}_x\text{N}_{1-\delta}$ ($x > 0$) and 40% in $\text{ScO}_x$. At lower $T_D$ the amount of scandium bonded to oxygen increases for all three substrates used. At a $T_D = 700$ °C, 24% of Sc is bonded to oxygen in films on $r$-plane $\text{Al}_2\text{O}_3$, while up to 40%–47% of Sc-O bonds are detected for the films on $c$-plane $\text{Al}_2\text{O}_3$ and $\text{MgO}(1 1 1)$ substrates. Figure 5(c) shows the evolution of the relative BE difference between the oxynitride and the nitride components in the Sc 2p core level spectra. The total amount of oxygen and the relative position of the oxynitride peak ($\text{ScO}_x\text{N}_{1-\delta}$ ($x > 0$)) in the Sc 2p core level vary with the temperature of deposition. The higher the oxygen content (the lower the temperature), the larger the shift of the oxynitride peak towards higher BE, which indicates that the O-to-N ratio in oxynitride increases.

The compositions of the ScN films as determined from XPS, are listed in table 2. Fluorine impurities were detected at around 1% or less. The anion/cation ratio varies from 1.06 for the films with the lowest content of oxygen to 1.23–1.34 for the films with the highest amount of oxygen. A higher $T_D$ results in thin film containing a low amount of oxygen (4%–5%) with a high N/Sc ratio between 0.93 and 0.97 for the film on $\text{MgO}(1 1 1)$ and $r$-plane $\text{Al}_2\text{O}_3$, respectively. The oxygen detected in those two films is mostly present as oxynitride. The film deposited on $c$-plane $\text{Al}_2\text{O}_3$ had a lower N/Sc ratio of 0.75 and contained a significant amount of oxygen, 16%, of which 10.3% is due to $\text{ScO}_x\text{N}_{1-\delta}$ ($x > 0$) and 5.6% due to an oxide. A lower $T_D$ yields ScN films containing higher content of oxygen (10%–15%), found as oxynitride and oxide and associated with lower ratio of N/Sc between 0.53 ($c$-plane $\text{Al}_2\text{O}_3$ substrate) and 0.79 ($r$-plane $\text{Al}_2\text{O}_3$ substrate). The XPS analysis of the surface of the films grown at 700 °C, and the one on $c$-plane $\text{Al}_2\text{O}_3$ grown at 820 °C revealed a high content of carbon even after a sputter cleaning process (which was kept identical for all samples). This amount of carbon is an indication of the effectiveness of the sputter cleaning, which is more difficult to achieve due to the high roughness and the surface morphology of the film, leading to shadowing on the significant fractions of the surface. Note here that the films presenting a high content of carbon on surface are also the ones with a contamination peak contribution in the O 1s core level spectra (see table 2 and figure 4, plus figure S2 in the supplementary information representing the C 1s core level spectra).

Figure 6(a) shows the electrical resistivity of the thin films measured by four-point probe at room temperature. Differences between the substrates used are noticeable. The lowest electrical resistivities (45 to 370 $\mu\Omega$ cm) were measured on films deposited on MgO(1 1 1), then higher electrical resistivities (55 to 625 $\mu\Omega$ cm) were measured on films grown on $r$-plane $\text{Al}_2\text{O}_3$. Finally, on $c$-plane $\text{Al}_2\text{O}_3$, higher resistivities are observed (180 to 2700 $\mu\Omega$ cm). On the three substrates, all films exhibited a decrease of the electrical resistivity when the $T_D$ increases from 700 °C to 950 °C or when the oxygen

![Figure 2. Top view SEM image/surface morphology of the ScN films deposited at different temperatures (700 °C, 820 °C and 950 °C) on: c-plane $\text{Al}_2\text{O}_3$, $\text{MgO}(1 1 1)$ and r-plane $\text{Al}_2\text{O}_3$.](image-url)
content into the film decreases. Figure 6(b) shows the Seebeck coefficient of the nine films. The films deposited on the three at the low $T_D$ exhibited similar Seebeck coefficients around $-27 \mu V K^{-1}$. At a $T_D = 950 ^\circ C$, all films exhibited higher absolute values of Seebeck coefficient around $-40 \mu V K^{-1}$.

**Discussion**

The XRD analyses confirmed the rock salt crystal structure with a lattice constant close to the bulk values (4.5013 Å/ICDD 030656286). All ScN films were grown epitaxially on the three substrates with a (1 1 1) orientation on $c$-plane $Al_2O_3$ and MgO(111) and with a (0 0 1) orientation on $r$-plane $Al_2O_3$. The differences on cell parameter values between the nine films and the bulk value may originate from composition variation. A (1 1 1) orientation for the cubic cell ScN films is expected on $c$-plane $Al_2O_3$ with an epitaxial relationship of $[\overline{1} \overline{1} 1]_{ScN} // [\overline{0} 0 1]_{Al_2O_3}$ and $[2 2 \overline{1}]_{ScN} // [1 0 \overline{0}]_{Al_2O_3}$. Epitaxial growth will be possible with a compressive stress on $c$-plane $Al_2O_3$ (mismatch: +14%) and MgO(111) (mismatch: +6%) and with a tensile/compressive stress for the film grown on $r$-plane $Al_2O_3$ (mismatch: −5%; +12%). A detailed scheme of the epitaxial relationship between the substrates and ScN is presented in the supplementary information. The increase of temperature increased the epitaxial quality of the film with an increase of the main diffraction peak and a reduce of the $\Delta \omega$ and $\Delta \phi$ associated to the main orientation of the film. At high temperature, the three films have an equivalent epitaxial quality (similar $\Delta \omega$ and $\Delta \phi$). The increase of the epitaxial quality is mainly due to the increase of energy brought during deposition by the temperature which increases the adatom mobility at the surface. As said before, the term epitaxial-like growth is used here to distinguish this from global epitaxy.

The morphology of the films at 950 °C varies depending of the substrates. This feature is highly correlated to the type of orientation and type of growth of the film. The morphology is also temperature-dependent on all three substrates with the suppression of mound grain growth when $T_D$ increases.

Figure 3. XPS peak models of Sc 2p and N 1s core level spectra acquired from films deposited on $c$-plane $Al_2O_3$, MgO(111), and $r$-plane $Al_2O_3$ at 700 °C, 820 °C and 950 °C.
The formation of this mound structure is commonly observed on Sn thin film and other transition metal nitrides [16, 20, 21, 25, 42–44]. Adatom mobility on a surface is affected by the presence of different defects or steps. The down-step motion of an adatom at a step edge is limited by the Ehrlich–Schwoebel barrier and favoring the uphill migration on terraces [45, 46]. Varying the barrier magnitude which depends of $T_D$, the kinetic surface roughness and faceting varies [47]. A high deposition temperature provides sufficient adatom mobility to suppress the surface roughening. As observed in this study and on all three substrates, an increase of temperature decreases the number of exaggerated grain growth to dense and homogenous films composed of nanometric size grains at 950 °C.

Figure 4. XPS peak models of O 1s core level spectra acquired from films deposited on c-plane $\text{Al}_2\text{O}_3$, MgO(1 1 1), and r-plane $\text{Al}_2\text{O}_3$ at 700 °C, 820 °C and 950 °C.

Figure 5. Summary of the Sc 2p XPS peak modelling performed for films grown on c-plane $\text{Al}_2\text{O}_3$, MgO(1 1 1) and r-plane $\text{Al}_2\text{O}_3$ and at 700 °C, 820 °C and 950 °C. (a) Fraction of the scandium in ScO$_{x}$N$_{y}$ + ScO$_{x}$; (b) fraction of oxygen detected as an oxide ScO$_{x}$; (c) binding energy (BE) difference between the ScN and the ScO$_{x}$N$_{y}$ peaks in the Sc 2p spectra.
Along with the improvement of the epitaxial quality when the \( T_D \) was increased from 700 °C to 950 °C, the nitrogen content and/or the ratio N/Sc increased from 0.75 to 0.97. The reactivity of scandium with nitrogen is enhanced at higher temperature as usually observed in DC-magnetron sputtering of nitride materials [48]. In the present case, a temperature of 950 °C leads to deposition a film with a ratio close to 1:1 (Sc:N) when grown on MgO(1 1 1) and r-plane Al\(_2\)O\(_3\).

By XRD, the presence of oxide is only detected on the low-temperature-grown films on c-plane Al\(_2\)O\(_3\) and on MgO(1 1 1). In contrast to the XRD with a Bragg-Brentano configuration where no oxide was detected, XPS revealed the presence of Sc\(_2\)O\(_3\) in the film grown on r-plane. This film has a higher amount of oxide detected than the other films grown at the same temperature. This apparent difference between XRD and XPS can be due to: (1) the low quantity of oxide (≈5%), (2) a possibility that this oxide is amorphous or oriented with a tilt relative to the normal of the surface and (3) the lower amount of oxide on the two other substrates was detected because of its preferential orientation (3 3 2) on c-plane sapphire and MgO (1 1 1). Note here that XPS is a surface analysis technique. A larger total amount of oxide in the film or at the substrate/film interface was not excluded to explain the presence of the oxide diffraction peak for the film grown on c-plane Al\(_2\)O\(_3\) and MgO(1 1 1) at 700 °C. Increasing the deposition temperature yields ScN films containing lower amounts of scandium oxynitride or oxide. The oxygen incorporation occurs mainly as dissolution into the ScN lattice and a few percent as an oxide, most likely at grain boundaries [20].

At a \( T_D \) of 950 °C, even with the base pressure of 10\(^{-7}\) Pa, the oxygen incorporation occurred during deposition, mainly due to the residual water in the chamber leading to few at.% of oxygen in the film. At that highest \( T_D \), less than 1 at.% of the oxygen detected is from an Sc\(_2\)O\(_3\) oxide and 3%–5% of oxygen detected dissolved in ScN cell (as oxynitride) for the film grown on MgO(1 1 1) and r-plane Al\(_2\)O\(_3\). Thus, the films deposited on MgO(1 1 1) and r-plane Al\(_2\)O\(_3\) have similar impurities level and N/Sc ratio. The (1 1 1) oriented film epitaxially grown on c-plane Al\(_2\)O\(_3\) had different level of impurities with a higher oxygen content and lower N/Sc ratio. Similar observations can be made for the film grown at 820 °C and at 700 °C, where the film grown on c-plane sapphire had the highest amount of oxygen impurities. This difference may originate from the type of epitaxial-like growth between the films. In one case (on c-plane sapphire), twin domain growth occurred but in the other one (on MgO(1 1 1)). The presence of twin domain observed on ScN(1 1 1)/c-plane sapphire can be affiliated to defects/steps where adatom mobility (surface energy) is modified and favors the insertion of impurities (here oxygen). This phenomenon has been observed, for example, on a crystal of MgAl\(_2\)O\(_4\) where impurities (Si, Ti) insertion arises at the twin boundaries of the monocrystal [49].

The difference in oxygen content in the two films oriented (0 0 1) and (1 1 1) on MgO may originate from the orientation of the film and the surface terminated during growth, where, in the case of ScN, a (0 0 1) terminated surface will be less favorable to insertion of oxygen (impurities) compared to a (1 1 1) terminated surface. A high percentage of oxygen leads to the formation of oxide into the film located most likely at the grain boundary and defects and may deteriorate the overall electrical properties of the films.

The electrical resistivity is decreased by a factor 10 when the \( T_D \) is increased from 700 °C to 950 °C, and the absolute Seebeck coefficient values measured at room temperature are increased by 40%. Different hypotheses can be proposed to explain the improvement of the electrical properties with the temperature: (1) a densification of the film along with a morphology evolution; (2) an increase of the N/Sc ratio towards 1 along with a decrease of oxygen impurities (oxide/oxynitride), (3) the increase of the epitaxial orientation quality of the film.

At the highest \( T_D \) (950 °C), differences between substrates are only noticeable on the composition of the film. Indeed, epitaxial quality, morphology, densification, were comparable between samples. The three films exhibited equivalent Seebeck coefficient (within the error bar) but different electrical resistivities. The film exhibiting a high electrical resistivity of about 200 \( \mu \)\Omega cm was the one deposited on c-plane Al\(_2\)O\(_3\). This electrical conductivity is equivalent to the values reported in the literature between 200–800 \( \mu \)\Omega cm (or \( \sigma = 1 \times 10^3 \) to \( 5 \times 10^3 \) \( \text{S} \text{ cm}^{-1} \)) [14–16, 21, 50]. The electrical resistivities of the two other films grown are reduced to values around 50 \( \mu \)\Omega cm (or \( \sigma = 2 \times 10^4 \) \( \text{S} \text{ cm}^{-1} \)) which is an improvement of 400%. The increase of the electrical conductivity of the films grown on MgO(1 1 1) and r-plane Al\(_2\)O\(_3\) was related to the growth mode without twin domain.
the ones reported in previous works (values from 1 to 2°C) [14, 15] to 3% decreased the electrical resistivity by a factor of 3.2 which gives an estimated power factor of $-40 \mu V K^{-1}$ which gives an estimated power factor of $3.2 \times 10^{-3} W m^{-1} K^{-2}$ at room temperature.

**Conclusion**

In conclusion, epitaxial-like growth of ScN rock-salt structure was obtained on three different substrates with a (1 1 1) orientation on c-plane Al$_2$O$_3$ and MgO(1 1 1) and a (0 0 1) orientation on r-plane Al$_2$O$_3$. Increasing the deposition temperature increased the crystal quality of the film and yielded smoother and denser films on all three substrates. XPS analysis allowed the detection of the presence of scandium oxynitride and scandium oxide. This is connected to the adatom mobility during the growth process affected by the deposition temperature and the presence or not of twin domains. Twin domains are induced by the orientation and nature of substrate. Higher oxygen contaminations was observed on ScN(1 1 1) c-plane sapphire with the presence of twin domain in contrary to others films ScN(1 1 1)/MgO(1 1 1) and ScN(001)/r-plane sapphire.

At the highest deposition temperature of 950 °C, where the films deposited on the three substrates differ by the amount of oxygen contaminant, a reduction the amount of oxygen impurities to 3% decreased the electrical resistivity by a factor of four while maintaining the Seebeck coefficient values. These samples exhibited low electrical resistivity for ScN films, with room-temperature values of 50 $\mu$Ω cm and a Seebeck coefficient values of $-40 \mu V K^{-1}$ which gives an estimated power factor of $3.2 \times 10^{-3} W m^{-1} K^{-2}$ at room temperature.

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**Conflicts of interest**

There are no conflicts of interest to declare.

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![Figure 6. (a) Room temperature electrical resistivity versus the growth temperature ($T_D$) of the ScN films deposited on: c-plane Al$_2$O$_3$, MgO(1 1 1) and r-plane Al$_2$O$_3$. (b) Estimated Seebeck coefficient at 50 °C versus the growth temperature ($T_D$) of the same films. Measurements performed at an average temperature of 50 °C with a $\Delta T = 47 ^\circ C$ and at atmospheric pressure.](image-url)
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