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Increased electromechanical coupling in $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$

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AlN is challenged as the material choice in important thin film electroacoustic devices for modern wireless communication applications. We present the promise of superior electromechanical coupling (k_t^2), in $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$ by studying its dielectric properties. $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$ ($0 \leq x \leq 0.3$) thin films grown by dual reactive magnetron sputtering exhibited low dielectric losses along with minor increased dielectric constant (ϵ). Ellipsometry measurements of the high frequency ϵ showed good agreement with density function perturbation calculations. Our data show that k_t^2 will improve from 7% to 10% by alloying AlN with up to 20 mol % ScN. © 2010 American Institute of Physics. [doi:10.1063/1.3489939]

The thin film electroacoustic technology and the use of wurtzite semiconductors (namely AlN) has established a sector within the piezoelectric industry, specifically through the release of the thin film bulk-acoustic resonator duplex filters for the use in mobile phones. AlN has proven to be a superior material due to high acoustic and dielectric quality and compatibility with the electronic circuit fabrication. However, a limiting factor of AlN, and hence the whole thin film electroacoustic technology as such, is its low electromechanical coupling factor (k_t^2) of 6%–7%, which is the determining factor for bandwidth in filters. To compensate for the low k_t^2 , complex multicomponent devices are needed which puts high fabrication demands and increased costs, as well as constraints on the performance of the filters. Therefore, corresponding materials are sought with improved electromechanical coupling (k_t^2).

An increased piezoelectric modulus (d_{33}) has experimentally been shown when alloying AlN with ScN.^{1,2} Recently, first-principle calculations for random solid solutions of $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$ confirmed this increase in d_{33} and related it to an increase in the piezoelectric constant (e_{33}) together with a decrease in the stiffness constant (C_{33}),³ both along the c -axis. The material k_t^2 can then be described as: $k_t^2 = e_{33}^2 / [(C_{33} + e_{33}^2 / \epsilon_{33}) \epsilon_{33}]$, where ϵ_{33} is the c -axis dielectric constant. Knowledge of the dielectric properties in $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$ is hence of given significance for predicting the k_t^2 and consequently the applicability of the material for electroacoustic applications.

In this paper, we present measurements of the relative dielectric constant (ϵ_r) together with calculations of k_t^2 . Along with ϵ_r also the dielectric dissipation (D) is presented which is a key factor to illustrate the dielectric behavior and strengthen the expectations of this material for low loss electroacoustic devices. dc-magnetron sputter epitaxy deposition has been employed to synthesize $\text{Sc}_x\text{Al}_{1-x}\text{N}$ films ($0.1 \leq x \leq 0.3$) using substrate temperature (T_s) of 400 °C. Good quality AlN was deposited at $T_s = 800$ °C for reference. The Sc-containing films deposited at $T_s > 400$ °C had detectable leakage currents and were not suitable for this study. A 200 nm thick epitaxial (111) TiN bottom electrode layer was de-

posited on (0001) sapphire prior to the (0001) $w\text{-ScAlN}$. Deposition of the alloy was then employed utilizing cosputtering from pure metallic Al and Sc targets. The metallic concentration was varied by the power ratio of the Al and Sc targets keeping the total power constant. Reactive sputter deposition was achieved in an atmosphere of Ar and N_2 at a total pressure of 0.33 Pa. The film thicknesses were typically 250 nm. Au contacts (diameter of 600 μm) with Cr adhesion layer were deposited on top. The 1 kHz–1 MHz impedance response of the Au/Cr/ScAlN/TiN structures was measured with LCR-meter [inductance (L), capacitance (C), and resistance (R)]. Composition was determined by elastic recoil detection analysis (ERDA). X-ray diffraction (XRD) was utilized for the structural analysis and it confirmed epitaxial growth for all studied Sc-concentrations. Figure 1 shows the XRD response for $\text{Sc}_x\text{Al}_{1-x}\text{N}$ films ($0.1 \leq x \leq 0.3$). Only the (0001) orientation could be found in the ScAlN films. The AlN reference sample did possess the best XRD response. XRD gave good agreement between the lattice parameters and theoretical expected values⁴ for $x \leq 0.2$. A deteriorated crystalline quality of the $x = 0.3$ sample due to initial phase decomposition of the solid solution,⁴ however, made it impossible to measure the lattice parameters for that composition.

Figure 2(a) shows ϵ_r extracted from the capacitance (C) of the Au/Cr/ScAlN/TiN structure as $\epsilon_r = Ct / (A\epsilon_0)$, where t , A , and ϵ_0 are the film thickness, electrode area, and vacuum permittivity, respectively. It is noted that the capacitance was well defined over the frequency range and that any interface capacitance was excluded since no significant influence was seen when comparing structures having different film thicknesses (250–500 nm).

There is a close to linear trend in the ϵ_r increase for $x = 0$ to 0.2 of 9.9 to 13.7, from which the $x = 0.3$ data point notably deviates. To further study the dielectric behavior, the electronic contribution of the dielectric response (ϵ_∞) has been extracted from spectroscopic ellipsometry measurements [Fig. 2(b)] by fitting the measurement data to a multilayer model including the surface roughness, as measured by atomic force microscopy. The electronic contribution to the dielectric tensor was then extracted from the low dispersion region at 1670 nm wavelength. The experimental values were compared to density-functional perturbation

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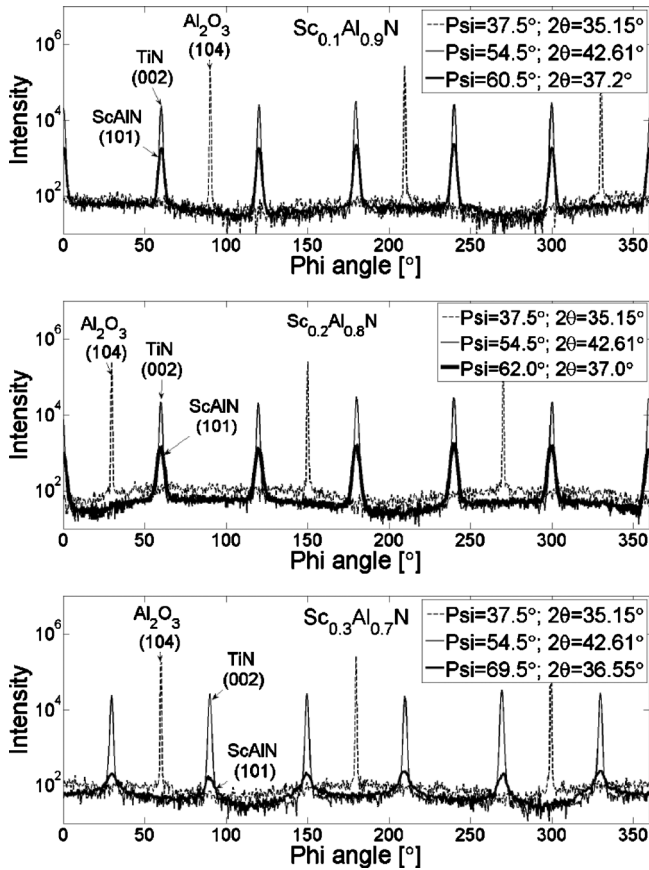


FIG. 1. X-ray diffractograms showing Phi-scans for $\text{Sc}_x\text{Al}_{1-x}\text{N}$ ($x=0.1, 0.2,$ and 0.3) films of the (104) Al_2O_3 , (002) TiN, and (101) $\text{Sc}_x\text{Al}_{1-x}\text{N}$ peaks.

theory (DFPT) (Ref. 5) calculations applied to supercells generated by the special quasirandom structure method³ [Fig. 2(b)]. As can be seen in Fig. 2(b) there is an increase also in ϵ_∞ due to the alloying. The experimental results follow the trend of the theoretical predictions well, but do possess lower values than predicted, as well as, have a slightly smaller difference between the ordinary and extraordinary directions of the crystal. Nevertheless, this agreement in ϵ_∞ also strengthens the applicability of the previously achieved theoretical results³ in this study. It is noted that the experimental

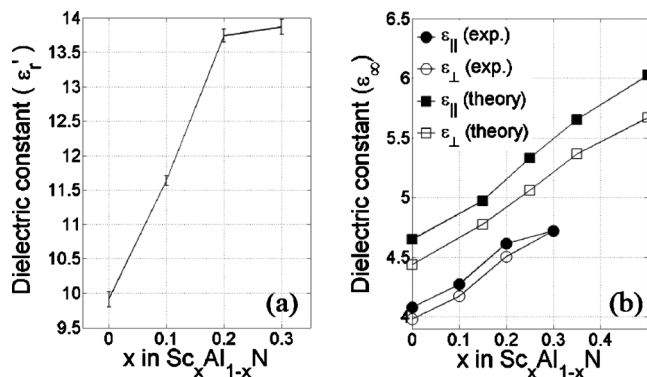


FIG. 2. (a) Relative dielectric constant (ϵ_r) for $\text{Sc}_x\text{Al}_{1-x}\text{N}$ ($0 \leq x \leq 0.3$) films. The values are obtained as an average of measurements at 1 kHz, 10 kHz, 100 kHz, and 1 MHz for three capacitive structures per sample. The error bars show the standard deviation. (b) Experimental results (exp.) of the high frequency dielectric constant obtained by ellipsometry for $\text{Sc}_x\text{Al}_{1-x}\text{N}$ along with DFPT calculations (theory) of the high frequency dielectric constant.

TABLE I. Measured relative dielectric constant (ϵ_r) values and dielectric loss (D), and theoretical values extracted from Refs. 3 and 9 of the stiffness (C_{33}) and piezoelectric constants (e_{33}) as well as calculated electromechanical coupling coefficients (k_t^2), $k_t^2 = e_{33}^2 / [(C_{33} + e_{33}^2 / \epsilon_{33}) \epsilon_{33}]$ for $\text{Sc}_x\text{Al}_{1-x}\text{N}$ ($0 \leq x \leq 0.3$) films.

Composition	ϵ_r	D	C_{33} (GPa)	e_{33} (C/m ²)	k_t^2 (%)
AlN	9.9	0.002	367 ^a	1.55 ^a	7.0
$\text{Sc}_{0.1}\text{Al}_{0.9}\text{N}$	11.6	0.005	303 ^b	1.60 ^b	7.5
$\text{Sc}_{0.2}\text{Al}_{0.8}\text{N}$	13.7	0.005	253 ^b	1.84 ^b	10.0
$\text{Sc}_{0.3}\text{Al}_{0.7}\text{N}$	13.9	0.008	211 ^b	2.12 ^b	15.0 ^c

^aReference 9.

^bReference 3.

^cFor the $\text{Sc}_{0.3}\text{Al}_{0.7}\text{N}$ it is noted that the crystal quality was highly degraded and deviation from theory could be seen giving higher uncertainty in the estimated electromechanical coupling.

AlN values of $\epsilon_\perp = 3.98$ and $\epsilon_\parallel = 4.08$, correspond rather well to earlier reported experimental results: $\epsilon_\perp = 4.13$ and $\epsilon_\parallel = 4.27$,⁶ $\epsilon_\perp = 4.05$ and $\epsilon_\parallel = 3.93$,⁷ and $\epsilon_\perp = 4.05$ and $\epsilon_\parallel = 4.32$.⁸ The measurement of the $x=0.3$ sample does not resolve any anisotropy which is correlated with the XRD results showing low crystalline quality for this sample. This lack of anisotropy causes the value to deviate slightly more from the theoretical predictions in the extraordinary direction than for the $x \leq 0.2$ samples, while the value in the ordinary direction still follows the trend. By comparing our ϵ_r -data to our ϵ_∞ -data [Figs. 2(a) and 2(b)] it is argued that the ϵ_r values extracted for $x \leq 0.2$ are not an underestimation of the ϵ_{33} while the value for $x=0.3$ could be an underestimation due to lost anisotropy. Thus, substituting the here determined ϵ_r for ϵ_{33} and utilizing the theoretically predicted values of C_{33} and e_{33} ,^{3,9} values for the k_t^2 in $\text{Sc}_x\text{Al}_{1-x}\text{N}$ could be calculated (Table I).

As can be seen in Table I, based on the measured dielectric constant values presented here, the k_t^2 in $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$ is predicted to have a nonlinear increase from 7% to 10% by alloying AlN with up to 20% ScN. Moreover, a significant further improvement in k_t^2 is indicated for ScN contents beyond 20%.

In summary, this study shows that $w\text{-Sc}_x\text{Al}_{1-x}\text{N}$, by its significantly increased electromechanical coupling coefficient and retained low dielectric losses, is a promising material choice for the AlN based thin film electroacoustic technology.

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