

# Supplementary Information: Thermal Conductivity and Optical Phonon Scattering of CMOS Compatible Aluminum Nitride-Based Ferroelectric Thin Films

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## S1. Film Growth and Characterization

$\text{Al}_{1-x}\text{B}_x\text{N}$  were grown varying the B concentration from  $x = 0, 0.01, 0.02, 0.06, 0.08, 0.13$ , and  $0.17$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films were grown varying the B concentration from  $x = 0, 0.02, 0.06, 0.08$ , and  $0.13$  and  $y = 0.20$ . Thin films are grown via reactive Radio Frequency (RF) magnetron co-sputtering from a metal Al and a ceramic BN target. Sc was deposited with a metal target and 200 W DC power.<sup>1</sup> One series of samples are approximately 200 nm thick.  $\text{Al}_{0.92}\text{B}_{0.08}\text{N}$  films from 13 nm to 199 nm were also investigated. Each film was grown on 100 mm diameter (100) highly As-doped (0.001–0.005 Ohm-cm) *n*-type Si substrates (Nova Electronic Materials). The chamber base pressures for depositions were below  $8 \times 10^{-8}$  Torr. Substrates were first loaded into the vacuum chamber and heated to 600 °C for 30 minutes then cooled down to 350 °C for another 30 minutes. A plasma treatment was performed at 20 mTorr in 20 sccm Ar and 20 sccm  $\text{N}_2$  with a 50 W substrate bias before the deposition. This step forms a  $\text{Si}_3\text{N}_4$  layer on the Si surface to improve AlN film texture and electrical properties.<sup>1</sup> The targets are cleaned in an Ar plasma for 5 minutes prior to all depositions. Deposition pressures are kept at 1.9 mTorr and a 24:16 Ar to  $\text{N}_2$  gas mixture for each composition. 2.5 W of substrate bias is utilized during each deposition to reduce formation of Grains Of Abnormal Texture (GOATs) and maintain smooth morphology.

We employed a PHI VersaProbe II X-ray Photoelectron Spectrometer (XPS) to quantify the B composition variation across the sample series<sup>1</sup>. Prior to XPS analysis, a 2-minute Ar sputter at 2 keV with a 2 mm  $\times$  2 mm spot size was performed to minimize contributions from surface oxidation and hydrocarbons. The Sc composition is determined.<sup>2</sup> Crystallinity and mosaic spread were characterized using a Panalytical Empyrean X-ray diffractometer (XRD) equipped with a Cu source and a five-axis cradle. All scans utilized a Bragg–Brentano HD incident optic and a PIXcel3D detector with programmable anti-scatter slits. Results are shown in Figs. S1(a) and (b) for  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films, respectively. An in-situ Film Sense Multiwavelength UV–Vis ellipsometer was used to measure refractive indices and verify film thickness as presented in Table S1.

Figure S1(c) presents the full width at half maximum (FWHM) rocking curve values of the corresponding samples, illustrating structural mosaicity increases and texture decreases monotonically with increasing B incorporation in both  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films. Atomic

force microscopy (AFM) topography scans comparing the surface of an as-received *n*-type Si substrate with that of a 200 nm-thick  $\text{Al}_{0.92}\text{B}_{0.08}\text{N}$  film are presented in Fig. S1(d). The root-mean-square (RMS) roughness values were determined to be 161 pm for the pristine Si surface and 380 pm for the  $\text{Al}_{0.92}\text{B}_{0.08}\text{N}$  film. All samples in this series are characteristically smooth except for  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  with high boron concentration.

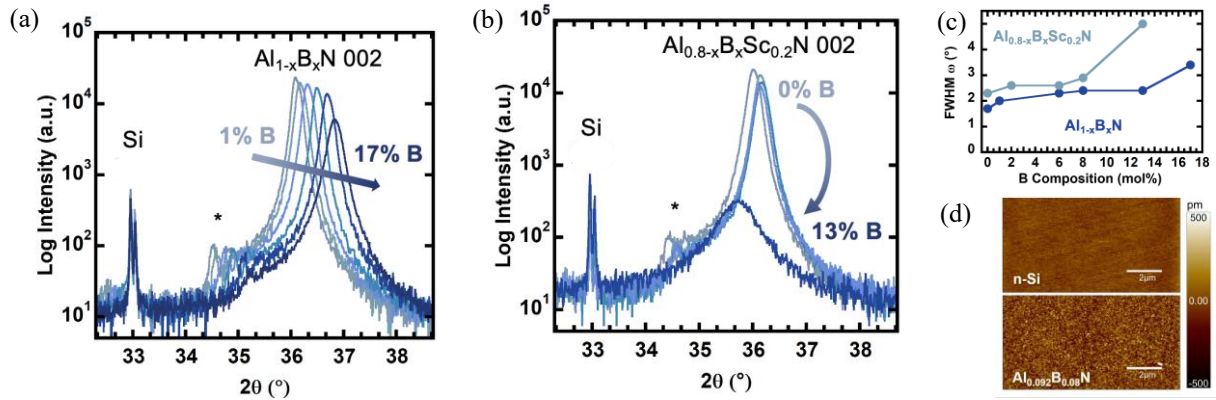


Figure S1. Offset XRD  $2\theta$ - $2\theta$  scans of (a) 200 nm 002  $\text{Al}_{1-x}\text{B}_x\text{N}$  peak where  $x = <0.01, 0.02, 0.06, 0.08, 0.13$ , and  $0.17$ , (b) 200 nm (002)  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  peak where  $x = <0.01, 0.02, 0.06, 0.08$ , and  $0.13$  and  $y = 0.20$ . (c) rocking curve FWHM of the  $2\theta$  002 peak as a function of B concentration. (d) AFM of as received *n*-type Si wafer surface roughness (top) in addition to 200 nm  $\text{Al}_{0.92}\text{B}_{0.08}\text{N}$  surface roughness (bottom). Starred peaks in (a) and (b) represent x-ray spectral lines.

Table S1. Film thicknesses measured by XRR

| Film                                                      | $d$ (nm) |
|-----------------------------------------------------------|----------|
| AlN                                                       | 195      |
| $\text{Al}_{0.99}\text{B}_{0.01}\text{N}$                 | 201      |
| $\text{Al}_{0.98}\text{B}_{0.02}\text{N}$                 | 198      |
| $\text{Al}_{0.94}\text{B}_{0.06}\text{N}$                 | 197      |
| $\text{Al}_{0.92}\text{B}_{0.08}\text{N}$                 | 202      |
| $\text{Al}_{0.87}\text{B}_{0.13}\text{N}$                 | 201      |
| $\text{Al}_{0.83}\text{B}_{0.17}\text{N}$                 | 213      |
| $\text{Al}_{0.80}\text{Sc}_{0.20}\text{N}$                | 203      |
| $\text{Al}_{0.78}\text{B}_{0.02}\text{Sc}_{0.20}\text{N}$ | 215      |
| $\text{Al}_{0.74}\text{B}_{0.06}\text{Sc}_{0.20}\text{N}$ | 210      |
| $\text{Al}_{0.72}\text{B}_{0.08}\text{Sc}_{0.20}\text{N}$ | 216      |
| $\text{Al}_{0.67}\text{B}_{0.13}\text{Sc}_{0.20}\text{N}$ | 214      |

## S2. P–E Loop Characterization

Polarization hysteresis measurements were carried out with an aixACCT TF 3000 Analyzer in combination with a 150 V High Voltage Amplifier. Triangular bipolar waveforms are used at 10 Hz, 200 Hz, and 1 kHz, respectively. 100 nm circular W top electrodes are sputtered using 100  $\mu\text{m}$  diameter shadow masks. Optical microscopy is used to determine relative electrode size for each electrical measurement. The highly doped *n*-type Si substrate acts as the bottom electrode in each measurement. The results for  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films are presented in Figs. S2 (a) and (b) respectively.

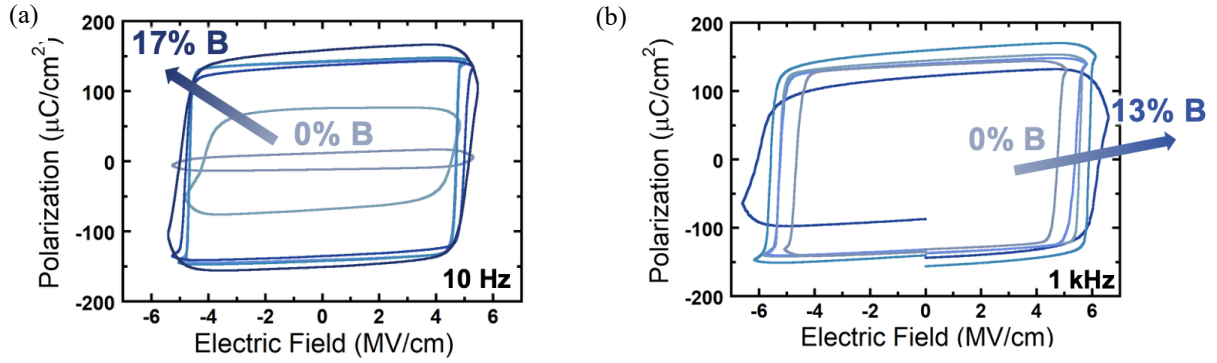


Figure S2. Polarization hysteresis progression of (a) 200 nm  $\text{Al}_{1-x}\text{B}_x\text{N}$  increasing B composition of  $x \sim 0.01, 0.02, 0.06, 0.08, 0.13$ , and 0.17 mol %; and (b) 200 nm  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  where  $x = < 0.01, 0.02, 0.06, 0.08$ , and 0.13 and  $y = 0.20$ .

### S3. Thermal Conductivity Measurement

#### S3.1 Time-Domain Thermoreflectance

The thermophysical properties of  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films were measured at room temperature and at elevated temperatures up to 400 °C by a time-domain thermoreflectance (TDTR) technique. The TDTR system employs a Ti:Sapphire oscillator (Spectra Physics Tsunami) with a central wavelength of approximately 808 nm, operating at an 80 MHz repetition rate. The laser is split into a pump beam and a probe beam and are focused down on the surface samples with  $1/e^2$  radii of  $\sim 10$  and  $\sim 5.5$   $\mu\text{m}$ , respectively. The pump beam is modulated at 8.4 MHz frequency using an electro-optical modulator (EOM), producing a periodic temperature rise on the sample's surface that alters reflectivity. the changes in reflectivity via the changes in probe beam reflectivity are detected using a balanced photodetector and a lock-in amplifier. More details on this measurement technique can be found in Refs.<sup>3,4</sup>

High temperature thermal conductivity measurements were performed with an INSTEC Vacuum Thermal Plate (HCP421V), with the temperature ramp set to 10 °C/min. The thermal conductivity of the substrate was independently measured at each temperature point. Leveraging this known quantity, thermal conductivity ( $\kappa$ ) and interfacial thermal resistance ( $R_{\text{Al}/\text{Al}_{1-x}\text{B}_x\text{N}} \text{ \& } \text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$ ) of the  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films were extracted. The thermal conductivity of  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films is determined by fitting a radially symmetric, multilayer heat diffusion model to the ratio of the in-phase and out-of-phase signal,  $V_{\text{in}}/V_{\text{out}}$ . Parameters used in TDTR data fitting are provided in Table S2, and films thicknesses are provided in Table S1.

Table S2. Thermal properties for TDTR data fitting process for  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films grown on n-type Si.

| Parameter/Layer                                                                                                                                           | Al (transducer)                                                  | $\text{Al}_{1-x}\text{B}_x\text{N}/\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$ (film) | Si (substrate)                   |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------|-------------------------------------------------------------------------------------------|----------------------------------|
| $d$ (nm)                                                                                                                                                  | measured with picosecond acoustics                               | measured by XRR                                                                           | bulk                             |
| $C_v$ ( $\text{MJ m}^{-3} \text{ K}^{-1}$ )                                                                                                               | Ref. <sup>5</sup>                                                | interpolated from Refs. <sup>6,7</sup>                                                    | Ref. <sup>5</sup>                |
| $\kappa$ ( $\text{W m}^{-1} \text{ K}^{-1}$ )                                                                                                             | measured with 4-point probe and Wiedemann Franz law <sup>8</sup> | measured with TDTR                                                                        | measured with TDTR independently |
| $R_{\text{Al}/\text{Al}_{1-x}\text{B}_x\text{N}} \text{ \& } \text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$ ( $\text{m}^2 \text{ K GW}^{-1}$ )           |                                                                  | Measured with TDTR                                                                        |                                  |
| $R_{\text{Al}/\text{Al}_{1-x}\text{B}_x\text{N}} \text{ \& } \text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}/\text{Si}$ ( $\text{m}^2 \text{ K GW}^{-1}$ ) |                                                                  | Ref. <sup>9</sup>                                                                         |                                  |

$d$  is the layer thickness,  $C_v$  is the volumetric heat capacity,  $\kappa$  is the thermal conductivity and  $R$  is the boundary resistance.

### S3.2 Uncertainty Analysis

The total uncertainty of the thermal conductivities are calculated using the following equation of total uncertainty ( $\varepsilon_i$ ):

$$\varepsilon_i = \sqrt{(\sigma_i^2) + \left(\sum_i \Delta_i^2\right)}$$

Here  $\sigma_i$  represents the standard deviations among multiple measurements across various spots and  $\Delta_i$  denotes the uncertainty associated with an individual parameter of the thermal model. An uncertainty of +/- 3 nm was assumed for the transducer thickness. The uncertainty in Al thickness was propagated jointly with the measured sheet resistance when calculating the Al thermal conductivity through the Wiedemann–Franz relation. 5% uncertainty was assumed for the film's heat capacity ( $C_{v2}$ ) and 10% uncertainty was assumed for both the thermal boundary resistance between the film and substrate ( $R_{Al1-xBxN/Si}$  &  $R_{Al1-x-yBxScyN/Si}$ ) as well as the thermal conductivity of the substrate ( $\kappa_3$ ).

#### S4. Infrared Variable Angle Spectroscopic Ellipsometry

The ellipsometric data were obtained using an IR-VASE (IR-VASE Mark II, J.A. Woollam Company) across the spectral range of 500-5000  $\text{cm}^{-1}$  at room temperature. Measurements were conducted at two incident angles of 50° and 70° at room temperature and the incident angle of 60° at elevated temperature, with a resolution of 16  $\text{cm}^{-1}$ . For high-temperature measurements, a TSEL1000 gas-tight ellipsometry stage was employed in conjunction with Linkam's NEXUS software (Windows platform). An inert argon atmosphere was continuously purged through the chamber to minimize oxidation. The temperature ramp rate was maintained at 25 °C/min. Following the high-temperature experiments, all samples were re-measured at room temperature. Infrared spectra collected before and after the ellipsometric measurements revealed no discernible differences, confirming that the samples did not undergo extensive oxidation, structural modification, or any other irreversible alteration as a consequence of the thermal cycling. The effective phonon lifetimes in the  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  were extracted by fitting a multi-oscillator dielectric function model to the complex, frequency-dependent ellipsometric response as presented in Table S3 and S4, respectively. In constructing the optical model, the underlying substrate was represented as bulk *n*-type silicon capped with an approximately 4 nm  $\text{Si}_3\text{N}_4$  interfacial layer. To account for morphological non-idealities, a surface roughness layer was also incorporated into the model, with best-fit values converging to roughness of up to ~5 nm. The fidelity of the fits was evaluated against the experimentally measured ellipsometric parameters ( $\Psi$  and  $\Delta$ ), ensuring that the extracted phonon lifetimes faithfully capture the intrinsic vibrational dynamics of the films.

The measured ellipsometric values ( $\Psi$  and  $\Delta$ ) represent the amplitude ratio and phase difference of the complex reflection coefficients ( $r_p$  and  $r_s$ ) of the sample for *p*- and *s*-polarized light, expressed as  $\rho = r_p/r_s = \tan \Psi e^{i\Delta}$ . A multi-layer optical model was constructed based on the measured ellipsometric data to extract the complex frequency-dependent dielectric function of each layer within the sample stack. In order to characterize the dielectric function,  $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$ , the data were modeled using Lorentz oscillators. Parameters for each oscillator model (including amplitude, energy centroid, and broadening) were optimized to align the model with the collected ellipsometric data by minimizing the mean square error. Finally, the effective phononic lifetime ( $\tau$ ) was derived from the relaxation rate, given as  $\tau = \Gamma^{-1}$ . The phonon-lifetime

uncertainties were determined using a uniqueness analysis, with the bounds defined by the damping-parameter values that increased the mean squared error by 10% from its minimum.

Table S3. Fitted model using Lorentzian oscillators for  $Al_{1-x}B_xN$

| <b>Al<sub>0.99</sub>B<sub>0.01</sub>N</b> |                  |                                     |                                     |
|-------------------------------------------|------------------|-------------------------------------|-------------------------------------|
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 2516.6           | 667.13                              | 16.000                              |
| <b>Extraordinary</b>                      | 2968.4           | 600.00                              | 13.305                              |
| <b>Al<sub>0.98</sub>B<sub>0.02</sub>N</b> |                  |                                     |                                     |
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 2454.8           | 667.79                              | 24.144                              |
| <b>Extraordinary</b>                      | 2858.5           | 625.00                              | 18.433                              |
| <b>Al<sub>0.94</sub>B<sub>0.06</sub>N</b> |                  |                                     |                                     |
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 2447.0           | 675.76                              | 38.918                              |
|                                           | 132.11           | 562.3                               | 38.75                               |
| <b>Extraordinary</b>                      | 2444.1           | 650.00                              | 34.136                              |
| <b>Al<sub>0.92</sub>B<sub>0.08</sub>N</b> |                  |                                     |                                     |
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 2291.3           | 687.21                              | 53.257                              |
|                                           | 177.26           | 566.95                              | 43.681                              |
| <b>Extraordinary</b>                      | 2313.5           | 675.00                              | 52.739                              |
| <b>Al<sub>0.87</sub>B<sub>0.13</sub>N</b> |                  |                                     |                                     |
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 2107.6           | 697.24                              | 66.362                              |
|                                           | 125.78           | 572.78                              | 38.551                              |
| <b>Extraordinary</b>                      | 2219.0           | 700.00                              | 70.440                              |
| <b>Al<sub>0.83</sub>B<sub>0.17</sub>N</b> |                  |                                     |                                     |
|                                           | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                           | 1864.1           | 705.00                              | 80.508                              |
|                                           | 85.295           | 576.01                              | 35.792                              |
| <b>Extraordinary</b>                      | 1742.3           | 725.00                              | 101.63                              |

Table S4. Fitted model using Lorentzian oscillators for  $Al_{1-x-y}B_xSc_yN$

| <b>Al<sub>0.80</sub>Sc<sub>0.20</sub>N</b>                 |                  |                                     |                                     |
|------------------------------------------------------------|------------------|-------------------------------------|-------------------------------------|
|                                                            | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                                            | 3123.9           | 632.47                              | 62.222                              |
|                                                            | 86.363           | 478.87                              | 54.194                              |
| <b>Extraordinary</b>                                       | 2409.7           | 600.00                              | 23.501                              |
| <b>Al<sub>0.78</sub>B<sub>0.02</sub>Sc<sub>0.20</sub>N</b> |                  |                                     |                                     |
|                                                            | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                                            | 2935.5           | 639.66                              | 83.508                              |
|                                                            | 132.64           | 499.28                              | 67.833                              |
| <b>Extraordinary</b>                                       | 2433.3           | 625.00                              | 38.796                              |
| <b>Al<sub>0.74</sub>B<sub>0.06</sub>Sc<sub>0.20</sub>N</b> |                  |                                     |                                     |
|                                                            | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                                            | 2788.4           | 645.41                              | 91.797                              |
|                                                            | 60.589           | 517.92                              | 77.243                              |
| <b>Extraordinary</b>                                       | 1847.7           | 650.00                              | 53.080                              |
| <b>Al<sub>0.72</sub>B<sub>0.08</sub>Sc<sub>0.20</sub>N</b> |                  |                                     |                                     |
|                                                            | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                                            | 2636.3           | 661.21                              | 113.50                              |
|                                                            | 264.00           | 529.35                              | 133.97                              |
| <b>Extraordinary</b>                                       | 1693.6           | 675.00                              | 67.978                              |
| <b>Al<sub>0.67</sub>B<sub>0.13</sub>Sc<sub>0.20</sub>N</b> |                  |                                     |                                     |
|                                                            | <b>Amplitude</b> | <b>Wavenumber (cm<sup>-1</sup>)</b> | <b>Broadening (cm<sup>-1</sup>)</b> |
| <b>Ordinary</b>                                            | 2235.2           | 683.30                              | 153.11                              |
|                                                            | 471.93           | 564.44                              | 163.57                              |
| <b>Extraordinary</b>                                       | 1576.5           | 700.00                              | 94.391                              |

## S5. Sound Speed Measurement

The longitudinal speed of sound was extracted from optical pump-probe picosecond acoustic measurements using the same two-tint time-domain thermoreflectance system described above. As the output of a Ti:sapphire oscillator (100 fs pulses, 80 MHz) is energetically separated into high-energy pump and low energy probe paths, the electro-optically modulated pump at 8.4 MHz creates a periodic heating event at the surface of the samples coated with aluminum. This heating event also generates a strain pulse that propagates through the film stack, partially transmitting/reflecting at the interfaces between the layers. The reflectivity of the aluminum transducer is interrogated using the probe, which is mechanically delayed following the incident pump pulse. Because the reflectivity of aluminum is proportional to both temperature and strain, signatures at short pump/probe delay times are indicative of the acoustic propagation times within each layer. Acoustic responses for the ~200 nm-thick crystallized  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  films are shown in Figs. S3 (a) and (b), respectively, where signatures from the Al/film and film/Si interfaces are identified.

The sign of the variation is determined as follows: when the acoustic pulse reflects off an interface with a layer of lower (or higher) acoustic impedance,  $Z$ , it undergoes a phase shift of  $\pi$  (or zero, respectively). If the acoustic impedances on either side of the interface are denoted by  $Z_1$  and  $Z_2$ , the reflection coefficient for normal incidence is given by  $R = (Z_2 - Z_1)/(Z_2 + Z_1)$ , where  $-1 < R < 1$ . The amplitude of the acoustic peaks in the thermoreflectance signal is influenced by the value of  $R$ , as well as the absorption and transmission coefficients of the layers that the acoustic echo traverses on its way back to the surface of the Al layer.

The acoustic impedance is defined as  $Z = \rho v$ , where  $\rho$  is the density and  $v$  is the longitudinal speed of sound in the layer. Since the sample geometry is as follows: Al/ $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}/\text{Si}$ , the strain wave undergoes null phase change at Al/ $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  interface, so the first echo peak is positive. A portion of the strain wave is propagated through the  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}$  and undergoes a  $\pi$  phase change at  $\text{Al}_{1-x}\text{B}_x\text{N}$  and  $\text{Al}_{1-x-y}\text{B}_x\text{Sc}_y\text{N}/\text{Si}$  interface, therefore the second echo peak is negative. Acoustic impedance of each layer is listed in the Table S5 shown below:

Table S5. Acoustic impedance of each layer in  $Al_{1-x}B_xN/Al_{1-x-y}B_xSc_yN$  films.

| Parameter/Layer                               | Al (transducer)    | $Al_{1-x}B_xN/Al_{1-x-y}B_xSc_yN$<br>(film) | Si (substrate)      |
|-----------------------------------------------|--------------------|---------------------------------------------|---------------------|
| density ( $g\ cm^{-3}$ )                      | 2.7                | 3.26                                        | 2.33                |
| longitudinal speed of sound ( $nm\ ps^{-1}$ ) | 6.42               | $\sim 10$ -11                               | 8.33*               |
| impedance (Rayl)                              | $17.3 \times 10^6$ | $\sim 33$ - $35.5 \times 10^6$              | $19.09 \times 10^6$ |

\*From ref. <sup>15</sup>

To accurately measure the longitudinal speed of sound in the cross-plane direction, the film thicknesses reported in Table S1 was used. The time intervals corresponding to the acoustic echoes from the  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN$  and  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN/Si$  interfaces recorded from the data shown in Figs S3(a) and (b) for  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN$ , respectively. The echo from the Al/ $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN$  interface appears at time  $t_1$ , and the echo from the  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN/Si$  interface arrives at time  $t_2$ . The cross-plane longitudinal sound speed in the film is then given by  $v = 2d / (t_2 - t_1) = 2d / \tau$ .

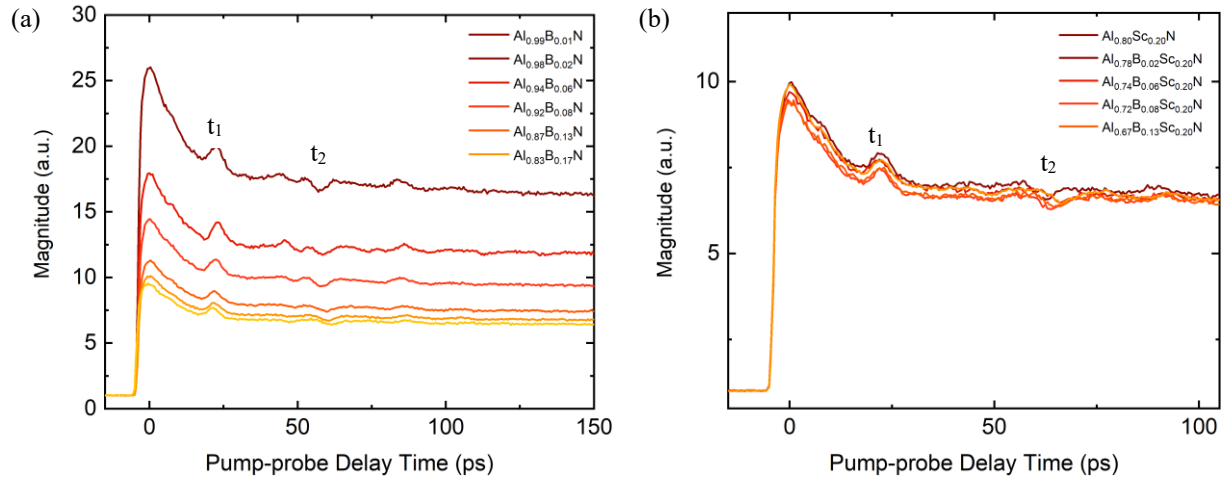


Figure S3. Picosecond acoustics measurements for (a)  $Al_{1-x}B_xN$  and (b)  $Al_{1-x-y}B_xSc_yN$  films deposited on n-type Si substrate. Arrival time of echoes from the  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN$  interface and  $Al_{1-x}B_xN$  and  $Al_{1-x-y}B_xSc_yN/Si$  interface are  $t_1$  and  $t_2$ . Cross-plane longitudinal speed of sound in the 3-component multilayer film is  $v = 2d / (t_2 - t_1)$ .

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