Supplemental materials

**Phonon Thermal Conductivity of Scandium Nitride for Thermoelectrics from First-Principles Calculations and Thin-Film Growth**

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Additional Scanning Electron Microscopy (SEM)

Fig. S1 shows SEM images of the surface morphology of all samples. As shown by XRD, the samples are highly textured polycrystalline ScN at deposition temperature \((T_d)\) up to 600 °C. This is especially easy to observe in the SEM image for the \(T_d = 400\) °C samples (Fig. S1(a)), with a rough surface with small triangular grains. At 700 °C deposition temperature, the film becomes smooth and fewer triangular facets, consistent with TEM that shows epitaxial growth.

FIG. S1 Top view SEM image of ScN films that were deposited at (a) \(T_d = 400\) °C, (b) \(T_d = 500\) °C, (c) \(T_d = 600\) °C, and (d) \(T_d = 700\) °C. (The samples with \(T_d = 400\) °C, \(T_d = 600\) °C, and \(T_d = 700\) °C were shown in the main article and are repeated here for convenience.)
Cross-section transmission electron microscopy

FIG. S2 Overview cross-section TEM image of ScN films that were deposited at (a) deposition temperature ($T_d$) = 400 ºC, (b) $T_d$ = 500 ºC, (c) $T_d$ = 600 ºC, and (d) $T_d$ = 700 ºC. (The samples with $T_d$ = 400 ºC, $T_d$ = 600 ºC, and $T_d$ = 700 ºC were shown in the main article and are repeated here for convenience.)

Fig. S2 shows the overview cross-section transmission electron microscopy (TEM) images of all ScN films. ScN deposited at $T_d$ = 400 ºC has a narrow columnar structure, again confirming that the $T_d$ = 400 ºC sample is polycrystalline, and highly textured as shown by XRD. For higher deposition temperatures of $T_d$ = 500-600 ºC, the films show an increase of the column width to ~70 nm from ~15 nm for $T_d$ = 400 ºC samples. Furthermore, they also show some degree of epitaxy at the initial state of growth – a few tens of nanometers – as seen by the triangular areas at the interface between film and substrate (see Fig. S2 (b), (c)). This initial epitaxy is not retained, as expected from competitive growth mechanisms at these moderate substrate temperatures.
FIG. S3 High resolution cross-section TEM image of the interface area between ScN film that was deposited at $T_d = 500 \, ^\circ$C and $\text{Al}_2\text{O}_3$ substrate.

A high-resolution TEM image of the substrate-film interface area for the film deposited at $T_d = 500 \, ^\circ$C is shown in Fig. S3. As seen in the image, several columns overlap each other; each column has initially nucleated and grown epitaxially on the substrate. Note that ScN films grown at $T_d = 500 \, ^\circ$C and $T_d = 600 \, ^\circ$C are similar in microstructure. When the deposition temperature was increased to $T_d = 700 \, ^\circ$C, the film has larger columnar width and shows epitaxial columns throughout the film thickness. The film evolution with deposition temperature is expected and explained by standard structure zone models.[1,2]

Fig. S4 shows cross-sectional high-resolution TEM images of the samples that were grown at $T_d = 400 \, ^\circ$C, $T_d = 500 \, ^\circ$C, and $T_d = 700 \, ^\circ$C. The film grown at the lowest deposition temperature exhibits domain formation due to secondary nucleation. The average size of these domains as observed in TEM is similar to that obtained from the XRD results. For higher deposition temperatures, these effects are less pronounced given the higher adatom mobility and correspondingly less secondary nucleation (see Fig. S4(b) and (c)).
FIG. S4 High resolution cross-section TEM images of ScN film deposited at (a) $T_d = 400 \, ^\circ$C, (b) $T_d = 500 \, ^\circ$C, and (d) $T_d = 700 \, ^\circ$C. The inset of each TEM image shows the corresponding electron diffraction. The $T_d = 600 \, ^\circ$C sample is not shown here since it has very similar in structure with the $T_d = 500 \, ^\circ$C sample.
Thermal conductance of the Al/ScN interfaces

The thermal conductance of Al/ScN was measured by time-domain thermoreflectance (TDTR) and is plotted in Figure S5. For the samples deposited at 400 °C, 500 °C, and 700 °C, the thermal conductance of the Al/ScN interface is 90 – 100 MW m⁻² K⁻¹. However, for the sample deposited at 600 °C, the thermal conductance is ~70 MW m⁻² K⁻¹. Since we deposited the Al film on all samples using the same chamber, the low thermal conductance might be due to a thicker oxide layer on the surface of the sample or lower quality (higher impurity content) of the sample.

FIG. S5 Thermal conductance of all ScN films
FIG. S6 Room temperature transport properties of ScN films that were deposited at $T_d = 400$ °C, 500 °C, 600 °C, and 700 °C, where (a) electrical resistivity and (b) Seebeck coefficient.

The electrical transport properties of all investigated ScN films are shown in Fig. S6. All samples have negative Seebeck coefficient indicating that electrons are majority charge carriers, as expected from previous studies.\[3,4\] The sample grown at $T_d = 400$ °C has the highest electrical resistivity in this set of samples, i.e., $61 \ \mu\Omega\text{m}$ with corresponding Seebeck coefficient of $-39 \ \mu\text{VK}^{-1}$. With increasing the decomposition temperature, the electrical resistivity tends to decrease, while the Seebeck coefficient decreases. This is associated with the observed microstructure shown in Figs. S1-S4, where the crystalline quality of the films
being improved with increasing deposition temperature. As a result, the sample grown at \( T_d = 700 \, ^\circ C \) show the lowest electrical resistivity of 4 \( \mu \Omega m \) and Seebeck coefficient of -30 \( \mu V K^{-1} \).

The Seebeck coefficients reported here are lower than in previous work,[3,4] because of the difference in oxygen impurities, since these samples were grown in high vacuum base pressure instead of ultra-high vacuum base pressure.
**Phonon self-energy**

It is generally assumed that the phonon self energy $\Sigma = \Delta + i \Gamma$ will shift the phonon frequencies $\omega$ by $\Delta$ and broaden the lines into Lorentzians with a FWHM of $\Gamma$. This is equivalent to approximating the real and imaginary part of the self-energy with constants. In general, that is not true, and the line shape is given by[5]

$$\sigma_{qq'}(\Omega) \propto \frac{2\omega_q \Gamma_q(\Omega)}{(\Omega^2 - \omega_q^2 - 2\omega_q \Delta_q(\Omega))^2 + 4\omega_q^2 \Gamma_q (\Omega)}.$$  

where $\Omega$ is the solid angle, $\omega_q$ is the phonon frequency, $q$ is the phonon wave vector, $s$ is the branch and the imaginary part of the self-energy ($\Gamma_q$) is given by

$$\Gamma_q(\Omega) = \sum_{s,s'} \frac{\hbar \pi}{16 (2\pi)^3} \int_{\mathbb{BZ}} |\Psi_{s's'}^{qsqs'}|^2 \times \left[ (n_{q',s' + 1}) \delta(\Omega - \omega_{q',s'} + \omega_{q,s}) + 2(n_{q',s} - n_{q',s'}) \delta(\Omega - \omega_{q',s'} + \omega_{q',s'}) \right] dq'dq'^*,$$  

where $\Delta_{qq'q''}$ ensures that $q + q' + q'' = K$, a reciprocal lattice vector and the three-phonon matrix elements is given by

$$\Psi_{ss'i}^{qq'q'} = \sum_{ijk} \sum_{\alpha\beta\gamma} \epsilon_{\alpha i}^{qs} \epsilon_{\beta j}^{qs'} \epsilon_{\gamma k}^{qs''} \times \Psi_{ijk}^{q} e^{i q r + i q' r' + i q'' r''},$$  

where $\epsilon_{\alpha i}^{qs}$ is the polarization vector, $ijk$ are the Cartesian indices, and $\alpha\beta\gamma$ are Cartesian indices. The real part of the self energy is given by Kramers-Kronig transformation:

$$\Delta(\Omega) = \frac{1}{\pi} \int \frac{\Gamma(\omega)}{\omega - \Omega} d\omega.$$
References


