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Characterizations of Seebeck coefficients and thermoelectric figures of merit for AlInN alloys with various In-contents

Jing Zhang, a) Hua Tong, Guangyu Liu, Juan A. Herbsommer, G. S. Huang, and Nelson Tansu b)
Center for Optical Technologies, Department of Electrical and Computer Engineering, Lehigh University, Bethlehem, Pennsylvania 18015, USA

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Thermoelectric properties of AlInN alloys, grown by metalorganic vapor phase epitaxy (MOVPE), with In-contents (x) from 11% up to 21.34% were characterized and analyzed at room temperature. The thermoelectric figure of merit (Z*T) values of the n-Al1-xInxN alloys were measured as high as 0.391 up to 0.532 at T = 300 K. The use of high In-content (x = 21.34%) AlInN alloys leads to significant reduction in thermal conductivity \([\kappa = 1.62 \text{ W/(mK)}]\) due to the increased alloy scattering, however, the optimized thermoelectric material was obtained for AlInN alloy with In-content of 17% attributed to its large power factor. © 2011 American Institute of Physics. [doi:10.1063/1.3553880]

I. INTRODUCTION

The high power density requirements in III-Nitride lasers and light-emitting diodes,1–15 transistors,16 and solar cells17–20 lead to the demand for solid state cooling technology,21–24 in particular for nitride-based alloy that can be integrated with GaN devices. III-Nitride alloys have shown promising thermoelectric figures of merit (Z’T),25–27 in particular for materials based on AlGaN28,29 and InGaN30,31 alloys. The thermoelectric properties for RF-sputtered AlInN had also been reported.27–30

Our recent works reported the high Z’T value for lattice-matched AlInN alloy grown by metalorganic vapor phase epitaxy (MOVPE).32 The investigation of thermoelectric characteristics of MOVPE-grown AlInN alloys is relatively lacking. Further investigations of the thermoelectric characteristics for MOVPE-grown AlInN alloys with various In-contents are of great interest for improved understanding of the important parameters necessary to further optimize the Z’T value in this material system.

In this work, we present the thermoelectric properties of AlInN alloys for various In-contents (x). The growths of n-AlInN alloys were carried out by MOVPE. The thermal conductivities (κ) of Al1-xInxN alloys were measured by 3ω differential method.33–35 The electrical conductivities (σ) and Seebeck coefficients (S) of the materials were measured by Hall and thermal gradient methods,36–38 respectively.

II. MOVPE EPITAXY OF ALINN ALLOYS

All the growths of AlInN films were performed on undoped-GaN (2.8 μm)/sapphire substrates. TMIn and TMAI were used as group III precursors, and NH3 was used as group V precursor. The growths of GaN template on sapphire substrate were performed by using 30-nm low temperature (Tg = 535 °C) GaN buffer, and then followed by high temperature (Tg = 1080 °C) GaN growth. The background n-type carrier concentrations of the GaN templates employed in the studies are 5 × 1016 cm–3. The growth temperatures of the n-Al1-xInxN alloys (∼200 nm thick) ranged between 750–790 °C with growth pressure of 20 Torr.

The In-contents (x) of Al1-xInxN epilayers were measured by X-ray diffractometer (XRD) for c-axis orientation, resulting in x = 0.11, 0.17, and 0.2134 (Fig. 1). The lattice-mismatch strain in the a-axis (Δa/a) between Al1-xInxN alloys and GaN as a function of In-content (x) are shown in Fig. 1. As the In-content in Al1-xInxN alloys reached ∼17% (Al0.83In0.17N), the Δa/a was measured as −0.18% which corresponded to nearly lattice-matched layer. For Al0.83In0.17N alloys, the growth temperature and V/III ratio were 780 °C and 9300, respectively. For Al0.83In0.11N and Al0.80In0.21N alloys, the growth temperatures were 790 °C and 750 °C, respectively.

Note that the increasing FWHM of the XRD rocking curves for AlInN with higher In-content can be attributed to the increasing phase separation in the film. Crack-free films were obtained for lattice-matched (x = 17%) and compressively-strained (x = 21.34%) samples, while cracks were observed tensile-strained samples (x = 11%).

FIG. 1. (Color online) The XRD rocking curves in c-axis for n-Al1-xInxN thin films grown on GaN/sapphire template.

a)Electronic mail: jiz209@lehigh.edu.

b)Electronic mail: tansu@lehigh.edu.
III. ELECTRICAL CONDUCTIVITIES AND SEEBECK COEFFICIENTS OF AlInN ALLOYS

The carrier mobilities of n-Al$_{1-x}$In$_x$N alloys were measured by the Hall method, as shown in Fig. 2. The background n-type carrier concentrations for two Al$_{0.83}$In$_{0.17}$N samples were measured as $5.1 \times 10^{18}$ cm$^{-3}$ and $1.6 \times 10^{18}$ cm$^{-3}$, respectively. For Al$_{0.89}$In$_{0.11}$N and Al$_{0.79}$In$_{0.21}$N alloys, the background n-type carrier concentrations were measured as $1.1 \times 10^{18}$ cm$^{-3}$ and $2.2 \times 10^{18}$ cm$^{-3}$, respectively. Note that the two lattice-matched Al$_{0.83}$In$_{0.17}$N samples were grown separately by MOVPE, and the difference in the background carrier concentration in the two samples can be attributed to the different impurity conditions from the epitaxy. Our finding is similar to the background carrier concentration range reported by others, which reported background doping in the range of $1-5 \times 10^{18}$ cm$^{-3}$. The difference in the background doping range for MOVPE-grown AlInN material is attributed to the nitrogen vacancies or residual oxygen impurities.

The carrier mobilities were measured as 290 cm$^2$/Vs and 462 cm$^2$/Vs for Al$_{0.83}$In$_{0.17}$N (n = $5.1 \times 10^{18}$ cm$^{-3}$) and Al$_{0.83}$In$_{0.17}$N (n = $1.6 \times 10^{18}$ cm$^{-3}$), respectively. The results show that higher carrier density leads to higher electrical conductivity and lower electron mobility. Despite its relatively low carrier concentration, the Al$_{0.89}$In$_{0.11}$N sample exhibits lower carrier mobility, which can be attributed to the cracks in the film. The cracks in the Al$_{0.89}$In$_{0.11}$N thin film is related to the dislocation density from the growths of tensile film on GaN template. With higher cracking density, the dominant crystal dislocation scattering reduces the electron mobility in AlInN alloy, which is in agreement with the simulation based on relaxation time approximation.

The Seebeck coefficients were determined by thermal gradient method, as illustrated in Fig. 3(a), which was similar to the method employed in references 36–38. When a temperature gradient was created in the sample [Fig. 3(a)], both the voltage difference and temperature difference were measured. A hotplate was used to create the high temperature. Two type K thermocouples were attached to the top surface of AlInN sample via indium (In) metal to measure temperature difference. The Seebeck voltage was collected from the positive chromel electrodes of the thermocouples at the same time. Figure 3(b) shows the measured Seebeck voltages as a function of the temperature difference for Al$_{1-x}$In$_x$N alloys with various In-content (x) from x = 0.11 up to x = 0.2134 at T = 300 K. The Seebeck voltages for all the Al$_{1-x}$In$_x$N samples show

FIG. 2. (Color online) The measured electron mobility for Al$_{1-x}$In$_x$N alloys with various In-contents (x) from x = 0.11 up to x = 0.2134 at T = 300 K.

FIG. 3. (Color online) (a) The setup for the thermal gradient method for Seebeck voltage measurements of AlInN samples, (b) Seebeck voltage as a function of the temperature difference, and (c) measured Seebeck coefficients for Al$_{1-x}$In$_x$N alloys with various In-contents (x) from x = 0.11 up to x = 0.2134 at T = 300 K.
good linearity with the measured temperature difference. Note that the measured Seebeck voltages in Fig. 3(b) refer to the total Seebeck coefficients of AlInN and chromel electrodes combined. The corresponding Seebeck coefficients for AlInN films need to be compensated by the Seebeck coefficient of chromel at room temperature (21.5 μV/K). The Seebeck coefficients of Al$_{1-x}$In$_x$N alloys are shown in Fig. 3(c). The absolute Seebeck coefficients for the AlInN alloys are relatively large, which are in the range of $-3.2 \times 10^{-4}$ V/K up to $-6.025 \times 10^{-4}$ V/K, with the highest value measured for n-Al$_{0.83}$In$_{0.17}$N alloy ($n = 5.1 \times 10^{19}$ cm$^{-3}$).

Figure 4 shows the measured power factors ($P = S^2$α) for Al$_{1-x}$In$_x$N alloys with $x = 0.11 - 0.2134$ at $T = 300$ K. The power factors for Al$_{0.83}$In$_{0.17}$N (n = 1.1 × 10$^{18}$ cm$^{-3}$) and Al$_{0.79}$In$_{0.21}$N (n = 2.2 × 10$^{18}$ cm$^{-3}$) were measured as 4.77 × 10$^{-4}$ W/(mK$^2$) and 2.11 × 10$^{-3}$ W/(mK$^2$), respectively. The higher power factors were obtained for the n-Al$_{0.83}$In$_{0.17}$N alloys, which were 8.64 × 10$^{-3}$ W/(mK$^2$) and 2.30 × 10$^{-3}$ W/(mK$^2$) for $n = 5.1 \times 10^{18}$ cm$^{-3}$ and $n = 1.6 \times 10^{18}$ cm$^{-3}$, respectively. The large power factors for the Al$_{0.83}$In$_{0.17}$N alloys are attributed to the large electrical conductivities and Seebeck coefficients. Note that both Seebeck coefficient and electrical conductivity have dependencies on carrier concentration, which in turn lead to the power factor variation with carrier concentration.

IV. THERMAL CONDUCTIVITIES CHARACTERIZATIONS OF AlInN ALLOYS

The thermal conductivities of AlInN films were measured by employing the $3\omega$ differential method and the details of this method used here were discussed in reference 38. Cross sectional schematic of four-probe $3\omega$ measurement setup for n-Al$_{1-x}$In$_x$N films grown on GaN/sapphire template prepared with SiO$_2$ insulation layer is shown in Fig. 5(a). The insulating layers of 200 nm SiO$_2$ were deposited by plasma-enhanced chemical vapor deposition on the AlInN/GaN/sapphire samples, and the metal heater contacts of 20 nm Ti/130 nm Au were deposited by using electron beam evaporator. The top microscope image of the four-probe $3\omega$ measurement setup for n-Al$_{1-x}$In$_x$N films was shown in Fig. 5(b).

In our $3\omega$ measurement setup, a digital lock-in amplifier SR830 was employed to supply the driving AC current (I$_{3\omega}$) with sweeping frequency ω and collect the voltage (V$_{3\omega}$) as well as the third harmonic voltage (V$_{3\omega}$) of the metal stripe. A digital multimeter HP 34401A was used to measure the current in order to obtain the metal heater resistance. All the $3\omega$ measurements were performed at room temperature. The $3\omega$ measurement setup was calibrated by measuring the thermal conductivities of sapphire and SiO$_2$ using differential and slope methods. For calibration purpose, the thermal conductivities of sapphire and SiO$_2$ (T = 300 K) were obtained as 41 W/(mK) and 1.1 W/(mK), respectively, in good agreement with reported values.

Both the measured voltage V$_{3\omega}$ and in-phase V$_{3\omega}$ of the undoped GaN reference sample and the n-Al$_{1-x}$In$_x$N samples at $T = 300$ K are shown in Fig. 6(a) (n-Al$_{0.83}$In$_{0.17}$N sample with $n = 5.1 \times 10^{18}$ cm$^{-3}$), Fig. 7(a) (n-Al$_{0.83}$In$_{0.17}$N sample with $n = 1.6 \times 10^{18}$ cm$^{-3}$), Fig. 8(a) (n-Al$_{0.79}$In$_{0.21}$N sample with $n = 2.2 \times 10^{18}$ cm$^{-3}$), and Fig. 9(a) (n-Al$_{0.89}$In$_{0.11}$N sample with $n = 1.1 \times 10^{18}$ cm$^{-3}$). The sweeping frequency of the driving current (ω/2π) ranged from 100 Hz to 1000 Hz, which insured the thermal penetration depth to be larger than the thickness of thin film while smaller than the thickness of the substrate.
measured as 1.62 W/(mK), 4.87 W/(mK), 4.31 W/(mK) and the undoped GaN reference sample and the n-Al

frequency in logarithm scale from 100 Hz to 1000 Hz for both

used for the growths of n-Al

range (from 100 Hz to 1000 Hz) was used to calculate the

plate reference samples correspond to identical templates

used for the growths of n-Al

ples. The average value of

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The thermal conductivities of Al

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N alloy was similar to that of

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rering. However, the low thermal conductivity for the

Al

N can be attributed to the cracking from the ten-

sile strain in the alloy.

V. THERMOELECTRIC FIGURES OF MERITS

OF AllN ALLOYS

The Z* T = P × T/K values for n-Al

N alloys at T = 300 K are shown in Fig. 11. The Z* T values (T = 300 K) for n-Al

N alloys were measured as high as 0.391 up to 0.532. The highest Z* T value (T = 300 K) was achieved as 0.532 for Al

N (n = 5.1 × 10^18 cm^{-3}). The Z* T values for Al

N (n = 2.2 × 10^18 cm^{-3}) and Al

N (n = 1.6 × 10^18 cm^{-3}) were 0.391, 0.160 and 0.089, respectively.

The thermal conductivities of Al

N with different carrier concentrations are measured as relatively similar, thus the variation of the Z* T values with carrier concentration is attributed to the carrier-concentration-dependent Seebeck and electrical conductivity parameters. The power factor P for Al

N alloy was similar to that of Al

N (n = 1.6 × 10^18 cm^{-3}), however its low thermal

Note that the V

and in-phase V

of the undoped GaN template on sapphire were measured as reference samples. To ensure consistency in the measurements, all the GaN template reference samples correspond to identical templates used for the growths of n-Al

N samples.

The temperature oscillation amplitude T_{ac} for the samples can be extracted from both the V

and V

by using the following relation:  

\[ T_{ac} = 2 \frac{V_{300}}{V_{100}} \frac{dT}{dR} R \]  

The temperature oscillation amplitudes T_{ac} as a function of frequency in logarithm scale from 100 Hz to 1000 Hz for both the undoped GaN reference sample and the n-Al

N samples at T = 300 K are shown in Fig. 6(b) (n-Al_{0.83}In_{0.17}N sample with n = 5.1 × 10^{18} cm^{-3}), Fig. 7(b) (n-Al_{0.83}In_{0.17}N sample with n = 1.6 × 10^{18} cm^{-3}), Fig. 8(b) (n-Al_{0.70}In_{0.30}N sample with n = 2.2 × 10^{18} cm^{-3}), and Fig. 9(b) (n-Al_{0.39}In_{0.11}N sample with n = 1.1 × 10^{18} cm^{-3}). Therefore, the temperature raise ΔT_{ac} was obtained by subtracting the T_{ac} of the undoped GaN reference sample from the n-Al

N samples. The average value of ΔT_{ac} for the entire frequency range (from 100 Hz to 1000 Hz) was used to calculate the thermal conductivity (κ) of the n-Al

N samples.

The measured thermal conductivities of Al

N alloys with x = 0.11–0.2134 at T = 300 K were shown in Fig. 10. The thermal conductivities of Al

N alloys were measured as 1.62 W/(mK), 4.87 W/(mK), 4.31 W/(mK) and 1.62 W/(mK) for x = 0.11, 0.17 (n = 5.1 × 10^{18} cm^{-3}), 0.17 (n = 1.6 × 10^{18} cm^{-3}) and 0.2134, respectively. The reduction in the thermal conductivity of Al

N with x = 0.2134 can be attributed to the increase in the alloy scattering. However, the low thermal conductivity for the Al

N can be attributed to the cracking from the tensile strain in the alloy.
conductivity from the increased alloy scattering led to higher $Z^*T$ value of 0.391. For $\text{Al}_{0.89}\text{In}_{0.11}N$ alloy, the high cracking density of the material led to a reduction of the power factor, which resulted in a lower $Z^*T$ value.

VI. SUMMARY

In summary, the thermoelectric properties of MOVPE-grown n-$\text{Al}_{1-x}\text{In}_x\text{N}$ ($x = 0.11–0.2134$) alloys are presented. The record $Z^*T$ values of the $\text{Al}_{1-x}\text{In}_x\text{N}$ alloys were measured as high as 0.391 up to 0.532 at $T = 300$ K, which show significant improvement from the RF-sputtered $\text{AlInN}$ ($Z^*T = 0.005$, $T = 300$ K)\textsuperscript{28} and MOVPE-grown $\text{InGaN}$ ($Z^*T = 0.08$, $T = 300$ K).\textsuperscript{36,37} The improvement observed from the MOVPE-grown $\text{AlInN}$ alloys can be attributed to the increase in the Seebeck coefficient and electrical conductivity resulting in higher power factor, in comparison to those measured from MOVPE-grown $\text{InGaN}$ and RF-sputtered $\text{AlInN}$. The use of high In-content ($x = 21.34\%$) $\text{AlInN}$ alloys leads to significant reduction in thermal conductivity [$\kappa = 1.62$ W/(mK)] due to the increased alloy scattering, however, the use of high In-content $\text{AlInN}$ alloys leads to slight reduction in power factor. To optimize the high $Z^*T$ value in $\text{AlInN}$ material system, it is important to employ crack-free $\text{AlInN}$ thin film with large carrier concentration in order to obtain high power factor, while minimizing the thermal conductivity by employing $\text{AlInN}$ alloy with In-content in the range of $x = 17\%–22\%$. The finding indicates that MOVPE-grown $\text{AlInN}$ alloy as excellent thermoelectric material for III-Nitride device integration.

FIG. 8. (Color online) Measured (a) voltage $V_x$ and in-phase $V_{3x}$, and (b) temperature oscillation amplitude ($T_{ac}$) as a function of frequency in logarithm scale for n-$\text{Al}_{0.79}\text{In}_{0.21}$N sample with $n = 2.2 \times 10^{18}$ cm$^{-3}$ and undoped GaN/sapphire reference sample at 300 K.

FIG. 9. (Color online) Measured (a) voltage $V_x$ and in-phase $V_{3x}$, and (b) temperature oscillation amplitude ($T_{ac}$) as a function of frequency in logarithm scale for n-$\text{Al}_{0.89}\text{In}_{0.11}$N sample with $n = 1.1 \times 10^{18}$ cm$^{-3}$ and undoped GaN/sapphire reference sample at 300 K.

FIG. 10. (Color online) The measured thermal conductivities for $\text{Al}_{1-x}\text{In}_x\text{N}$ alloys with various In-contents ($x$) from $x = 0.11$ up to $x = 0.2134$ at $T = 300$ K.
In-contents (x) from x = 0.11 up to x = 0.2134 at T = 300 K.

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