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High-temperature characteristics of Seebeck coefficients for AlInN alloys grown by metalorganic vapor phase epitaxy

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The Seebeck coefficients of AlInN alloys, grown by metalorganic vapor phase epitaxy, with In-contents (*x*) from 0.38% up to 21.34%, were characterized and analyzed from room temperature (T = 300 K) up to high temperature (T = 382 K). The Seebeck coefficients of the n-type AlInN alloys show significant enhancement at higher temperature up to 382 K, in comparison to those measured at room temperature. Large Seebeck coefficients ($602.0-1233.2 \mu$ V/K) were obtained for the lattice-matched Al_{0.83}In_{0.17}N alloy ($n = 5.1 \times 10^{18}$ cm⁻³) from T = 300 K up to T = 382 K. The improvement of Seebeck coefficients for the n-type AlInN alloys will lead to ~1.5-4 times improvement of the thermopower at higher temperatures. © 2011 American Institute of Physics. [doi:10.1063/1.3624761]

I. INTRODUCTION

High power density and high-temperature requirements in III-nitride based device technologies are of great importance for lasers,^{1–6} light-emitting diodes,^{7–22} transistors,²³ and solar cells.^{24,25} The use of active solid state cooling technology^{26,27} is of great importance for efficient thermal management in high power devices. The availability of IIInitride thermoelectric materials, which can be directly integrated with GaN device technology, has an important role for active thermal cooling and efficiency improvement in nitride-based high-power devices operating at high current density and high temperature.^{1-11,23-25} Promising thermoelectric figures of merit (Z^*T) have been reported for IIInitride materials,²⁸⁻⁴² in particular for materials based on AlGaN,^{36,37} InGaN,^{38,39} and AlInN^{40,41} alloys. The thermoelectric properties for RF-sputtered AlInN had also been reported.28-31

Specifically, recent works on InGaN alloys have been shown to have very promising Z^*T at room temperature.^{38,39} Pantha and co-workers have also reported the power factor and thermal conductivity of InGaN alloys at elevated temperature (T \sim 450 K), which demonstrated its promise as the new thermoelectric material.^{38,39} Recently, Pantha and coworkers have also extended their works into the use of (Er+Si)-doped AlInGaN with low In-content $\sim 10\%$ and Al-content ~14%, resulting in promising Z^*T values up to high temperature.⁴² The incorporation of (Er + Si)-dopants in the quaternary AlInGaN alloys lead to reduction in thermal conductivity by \sim 2.5-times, which leads to the very promising improvement in Z^*T values of this material.⁴² Our recent works^{40,41} reported high Z^*T value for AlInN alloys with various In-contents grown on GaN template by metalorganic vapor phase epitaxy (MOVPE) at room temperature (T = 300 K).

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In this work, we present the characterizations of the Seebeck coefficients for n-type $Al_{1-x}In_xN$ alloys (x = 0.0038-

0.2134) in the temperature range from room temperature

(T = 300 K) up to high temperature (T = 382 K). The See-

beck coefficients of the binary AlN and InN alloys were also

characterized, in order to provide comparison to those meas-

ured from the n-type AlInN ternary alloys. All the n-type

Al_{1-x}In_xN alloys with various In-contents, as well as the AlN

and InN alloys, were grown by MOVPE. The thermal gradi-

ent method was employed for the Seebeck coefficient mea-

surements, similar to the approaches presented in Refs. 38-42.

The Seebeck coefficients of the n-type Al_{1-x}In_xN alloys

show significant enhancement at higher temperature up to

T = 382 K compared to those measured at room temperature

(T = 300 K). The Seebeck coefficients show increasing trend

with higher Al-content in the n-type Al_{1-x}In_xN alloys. Large

Seebeck coefficients were obtained for the lattice-matched

 $Al_{0.83}In_{0.17}N$ alloy $(n = 5.1 \times 10^{18} \text{ cm}^{-3})$ from room temper-

ature up to higher temperatures. The significant enhancement

of Seebeck coefficients of the n-type Al1-xInxN alloys

(x = 0.0038 - 0.2134) can potentially lead to ~1.5-4 times

Our recent work^{40,41} reported the epitaxy and thermo-

electric characteristics of AlInN alloys with various In-

contents at room temperature (T = 300 K). All the n-type

AlInN films were grown by MOVPE on undoped-GaN

(thickness = 2.8 μ m; n_{background} ~5 × 10¹⁶ cm⁻³) template

on c-plane sapphire substrates. The growth temperatures of

the n-type Al_{1-x}In_xN alloys (\sim 200 nm thick) ranged between

750-860 °C with growth pressure of 20 Torr. The measured

In-contents (x) of $Al_{1-x}In_xN$ epilayers were x = 0.0038, 0.11, 0.17, and 0.2134, respectively. For $Al_{0.83}In_{0.17}N$ alloys,

which are lattice-matched to GaN, the growth temperature

improvement of the thermopower at higher temperatures.

II. EPITAXY AND THERMOELECTRIC

CHARACTERISTICS OF ALINN ALLOYS

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and V/III ratio were 780°C and 9300, respectively. For Al_{0.9962}In_{0.0038}N, Al_{0.89}In_{0.11}N, and Al_{0.79}In_{0.21}N alloys, the growth temperatures were 860°C, 790°C, and 750°C, respectively. The background carrier concentrations of n-type Al_{1-x}In_xN alloys were measured by the Hall method. The background n-type carrier concentrations for two lattice-matched Al_{0.83}In_{0.17}N samples were measured as 5.1×10^{18} cm⁻³ and 1.6×10^{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 of two respectively. The details of the growth conditions of the AlInN alloys were measured as 1.1×10^{18} cm⁻³ and 2.2×10^{18} cm⁻³, respectively. The details of the growth conditions of the AlInN alloys were presented in Ref. 41.

High Z^{*T} values were reported at T = 300 K for MOVPE-grown n-type AlInN alloys,^{40,41} which 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 lattice-matched ntype Al_{0.83}In_{0.17}N with carrier concentration $n = 5.1 \times 10^{18}$ cm⁻³. The Z^{*T} values of the MOVPE-grown Al_{1-x}In_xN alloys are significantly higher than the RF-sputtered AlInN $(Z^{*T} = 0.005, T = 300 \text{ K})^{33}$ and MOVPE-grown InGaN $(Z^{*T} = 0.08, T = 300 \text{ K})^{.38,39}$ The improvement observed from MOVPE-grown AlInN alloys are primarily attributed to the larger Seebeck coefficient and electrical conductivity resulting in higher power factor, in comparison to those measured from MOVPE-grown InGaN³⁸ and RF-sputtered AlInN.³³

Large Seebeck coefficients obtained from the MOVPEgrown AlInN alloys directly contributed to the enhancement of the thermopower and Z^*T values.⁴¹ Therefore, the characteristics of Seebeck coefficients of MOVPE-grown AlInN alloys are important in order to understand the contribution of Seebeck coefficients for improved Z^*T values. However, the investigation of thermoelectric characteristics of MOVPE-grown AlInN alloys at higher temperatures is relatively lacking, which play important roles in higher temperature thermoelectric characteristics for MOVPE-grown AlInN alloys at higher temperatures are of great interest for improved understanding of the improvement of Z^*T values at higher temperatures in this material system.

III. HIGH TEMPERATURE CHARACTERISTICS OF SEEBECK COEFFICIENTS FOR ALINN ALLOYS

The thermal gradient method, which is similar to the method employed in Refs. 38–42, was employed to determine the Seebeck coefficients, and the experimental setup is shown in Fig. 1. When a temperature gradient was created in the sample (Fig. 1), both the voltage difference and temperature difference were collected at the same time. Two type-K thermocouples were attached to the top surface of AlInN samples via indium (In) metal contacts to measure temperature difference. The positive chromel electrodes of the thermocouples were utilized to collect the Seebeck voltage, which was measured by a digital multimeter HP 34401A. The Seebeck voltages (ΔV) for all the Al_{1-x}In_xN samples show good linearity with the measured temperature difference. The total Seebeck coefficient was determined by using - ΔV /



FIG. 1. (Color online) The setup for the thermal gradient method for Seebeck voltage measurements.

 ΔT relation for all the n-type Al_{1-x}In_xN samples, which refer to the total Seebeck coefficients of n-type Al_{1-x}In_xN alloys combined with that of the chromel electrode. The corresponding absolute Seebeck coefficients for n-type Al_{1-x}In_xN films need to be compensated by the Seebeck coefficient of chromel (21.5 μ V/K) at room temperature (T = 300 K),⁴³ which changes minimally (~0.3 μ V/K)⁴⁴ for temperature range (T = 300–382 K) used in our experiments.

The measured Seebeck voltages (ΔV) as a function of the temperature difference (ΔT) for Al_{1-x}In_xN (x = 0.0038, 0.11) alloys from T = 300 K up to T = 382 K are shown in Figs. 2(a) and 3(a), respectively. The absolute Seebeck coefficients of $Al_{1-x}In_xN$ (x = 0.0038, 0.11) alloys at various temperatures are shown in Figs. 2(b) and 3(b), respectively. The absolute Seebeck coefficients for both Al_{1-x}In_xN (x = 0.0038, 0.11) tensile films show increasing trend with raising temperatures. For the Al_{0.9962}In_{0.0038}N sample with lowest In-content, the absolute Seebeck coefficients were measured as high as 960.3–1143.0 μ V/K for temperatures range from 300 K up to 382 K. For the Al_{0.89}In_{0.11}N sample, the absolute Seebeck coefficients were measured as 323.2-401.6 μ V/K for temperatures range from 300 K up to 382 K. Note that both Al_{0.9962}In_{0.0038}N and Al_{0.89}In_{0.11}N samples have cracks in the films, which are related to the dislocation density of the tensile films grown on GaN templates. The cracking of the tensile films will reduce the relaxation time of the carriers, which will lead to lower Seebeck coefficients of the films.⁴¹

Figures 4(a) and 4(b) show the measured Seebeck voltages (ΔV) as a function of the temperature difference (ΔT) and the absolute Seebeck coefficients for Al_{0.79}In_{0.21}N alloy from T = 300 K up to T = 382 K, respectively. As the Al_{0.79}In_{0.21}N sample was grown on GaN template with compressive strain, the thin film was crack-free. The absolute Seebeck coefficients for the compressive Al_{0.79}In_{0.21}N film show increasing trend with raising temperatures, which were measured as 386.4–460.7 μ V/K for temperatures range from T = 300 K up to T = 382 K.

Figures 5(a) and 5(b) show the measured Seebeck voltages (ΔV) as a function of the temperature difference (ΔT) for both lattice-matched 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) samples from





FIG. 2. (Color online) (a) Seebeck voltage as a function of the temperature difference, and (b) measured Seebeck coefficients for n-type $Al_{0.9962}$ $In_{0.0038}N$ alloy from T = 300 K up to T = 382 K.

T = 300 K up to T = 382 K, respectively. The absolute Seebeck coefficients for both Al_{0.83}In_{0.17}N ($n = 5.1 \times 10^{18} \text{ cm}^{-3}$) and Al_{0.83}In_{0.17}N ($n = 1.6 \times 10^{18} \text{ cm}^{-3}$) samples are shown in Fig. 5(c). Both the lattice-matched $Al_{0.83}In_{0.17}N$ samples exhibit large Seebeck coefficients for various temperatures. The absolute Seebeck coefficients were measured as high as 602.0-1233.2 µV/K and 446.3-592.3 µV/K from 300 K up to 382 K for $Al_{0.83}In_{0.17}N$ ($n = 5.1 \times 10^{18} \text{ cm}^{-3}$) and $Al_{0.83}$ In_{0.17}N ($n = 1.6 \times 10^{18}$ cm⁻³) samples, respectively. Therefore, higher background doping level of the lattice-matched Al_{0.83}In_{0.17}N sample shows larger enhancement of the Seebeck coefficient with increasing temperature. In addition, the lattice-matched Al_{0.83}In_{0.17}N sample with higher doping level exhibits larger Seebeck coefficient for the temperature range of 300 up to 382 K, which can be attributed to larger relaxation time of carriers that results from better crystalline epitaxial material quality.^{36,37,41} Further, larger Seebeck coefficient of the higher background doping level Al_{0.83}In_{0.17}N sample can also be attributed to the hopping conductance.²⁸ Specifically, as the hopping conductance leads to the decrease of resistivity for increasing carrier concentration, the Seebeck coefficient increases with higher carrier concentration.²⁸

Therefore, the high-temperature Seebeck coefficient characteristics of the n-type $Al_{1-x}In_xN$ alloys show that with higher In-content, the Seebeck coefficients are reduced for the n-type $Al_{1-x}In_xN$ thin films for the temperature ranges

FIG. 3. (Color online) (a) Seebeck voltage as a function of the temperature difference, and (b) measured Seebeck coefficients for n-type $Al_{0.89}In_{0.11}N$ alloy from T = 300 K up to T = 382 K.

from T = 300 K up to T = 382 K. The Al_{0.9962}In_{0.0038}N sample with the lowest In-content exhibits the largest absolute Seebeck coefficient at various temperatures. The absolute Seebeck coefficients of the lattice-matched Al_{0.83}In_{0.17}N samples are larger than those of the tensile Al_{0.89}In_{0.11}N alloy and the compressive Al_{0.79}In_{0.21}N alloy. Despite its lower In-content of the Al_{0.89}In_{0.11}N sample, the Seebeck coefficient is lower than the lattice-matched Al_{0.83}In_{0.17}N samples and the Al_{0.79}In_{0.21}N sample, which can be attributed to the cracking of the tensile film that will reduce the relaxation time of the carriers.

The Seebeck coefficients of the n-type $Al_{1-x}In_xN$ alloys show increasing trend with raising temperatures, which are significantly higher in comparison with those for RFsputtered AlInN (20–50 μ V/K, T=300-400 K)³³ and MOVPE-grown InGaN (150–180 μ V/K, T=300-400K).^{38,39} Therefore, the increase of Seebeck coefficients of the MOVPE-grown n-type $Al_{1-x}In_xN$ alloys at higher temperatures can potentially lead to ~1.5–4 times enhancement of the thermopower at higher temperature up to T = 382 K.

IV. SEEBECK COEFFICIENTS OF INN AND ALN BINARY ALLOYS

For comparison purpose, the Seebeck coefficients were characterized for InN and AlN binary alloys at various



FIG. 4. (Color online) (a) Seebeck voltage as a function of the temperature difference, and (b) measured Seebeck coefficients for n-type $Al_{0.79}In_{0.21}N$ alloy from T = 300 K up to T = 382 K.

temperatures as supplementary to the characterizations for the ternary n-type $Al_{1-x}In_xN$ thin films. The AlN binary alloy growth was performed by MOVPE on undoped-GaN (2.8 μ m)/sapphire substrate for the purpose of Seebeck coefficient characterization. TMAI was used as group III precursors, and NH₃ was used as group V precursor, and the V/III molar ratio was 2222. The growth temperature was 1075 °C with growth pressure of 100 Torr. The growth rate was measured as 3.7 nm/min, and the thickness of the AlN film was 190 nm.

The InN binary alloy was grown on undoped-GaN (2.8 μ m)/sapphire substrate by pulsed growth MOVPE method.⁴⁵ TMIn was used as group III precursors, and NH₃ was used as group V precursor, and the V/III molar ratio was 12460. The growth temperature for the growth of InN layer was 575 °C with growth pressure of 200 Torr, and the thickness of the n-type InN film was 200 nm with carrier concentration n $\sim 3.2 \times 10^{19}$ cm⁻³.

The Seebeck coefficients of the binary n-type AlN alloy were determined by the thermal gradient method at various temperatures range from T = 300 K up to T = 322 K. The measured Seebeck voltages (ΔV) as a function of the temperature difference (ΔT) for AlN alloy at various temperatures are shown in Fig. 6(a). The Seebeck voltages (ΔV) show good linearity with the measured temperature difference



FIG. 5. (Color online) (a) Seebeck voltage as a function of the temperature difference for n-type $Al_{0.83}In_{0.17}N$ (n = 5.1 × 10¹⁸ cm⁻³) alloy, (b) Seebeck voltage as a function of the temperature difference for n-type $Al_{0.83}In_{0.17}N$ (n = 1.6 × 10¹⁸ cm⁻³) alloy, and (c) measured Seebeck coefficients for n-type $Al_{0.83}In_{0.17}N$ (n = 5.1 × 10¹⁸ cm⁻³ and n = 1.6 × 10¹⁸ cm⁻³) alloys from T = 300 K up to T = 382 K.

 (ΔT) at various temperatures for the binary AlN sample. The measured absolute Seebeck coefficients of n-type AlN alloy after the correction from the Seebeck coefficient of chromel are shown in Fig. 6(b). The absolute Seebeck coefficients for the n-type AlN alloy were measured as high as 992.5 μ V/K up to 1004.7 μ V/K from room temperature up to T = 322 K. The measured Seebeck coefficients of the AlN binary alloy are larger than the Al_{1-x}In_xN (x = 11%- 21%) ternary alloys at similar temperatures. Further, the Seebeck coefficients of the AlN binary alloy are measured as very similar with those



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FIG. 6. (Color online) (a) Seebeck voltage as a function of the temperature difference, and (b) measured Seebeck coefficients for n-type AlN alloy from T = 300 K up to T = 322 K.

for $Al_{0.9962}In_{0.0038}N$ alloy with the lowest In-content at various temperatures, within the experimental error.

The measured Seebeck voltages (ΔV) for binary n-type InN alloy as a function of the temperature difference (ΔT) for InN alloy at various temperatures are shown in Fig. 7(a). The Seebeck voltages (ΔV) for the InN sample also show good linearity with the measured temperature difference (ΔT) at various temperatures from T = 300 K up to T = 382 K for the binary InN sample. The absolute Seebeck coefficients of ntype InN alloy were measured as 33.2 μ V/K up to 81.2 μ V/K for T = 300 K up to T = 382 K, after the correction from the Seebeck coefficient of chromel [Fig. 7(b)].

The Seebeck coefficients of the binary n-type InN alloy are found to be an order of magnitude lower than those of ntype Al_{1-x}In_xN (x=0.38%-21%) ternary alloys at various temperatures. In addition, the lower Seebeck coefficients of the InN samples can partially be attributed to the much higher doping concentration of the InN alloy (n ~ 3.2×10^{19} cm⁻³), which is in agreement with the theoretical simulation of the relationship between Seebeck coefficient and background carrier concentration.^{36,37}

For non-crack $Al_{1-x}In_xN$ alloys, the Seebeck coefficients are expected to be in the range between that of AlN and InN. For the low In-content $Al_{1-x}In_xN$ alloy (i.e., $x \sim 0.38\%$), the Seebeck coefficient is expected to be very similar to that measured for AlN alloy, which is in agreement with the com-

FIG. 7. (Color online) (a) Seebeck voltage as a function of the temperature difference, and (b) measured Seebeck coefficients for n-type InN alloy from T = 300 K up to T = 382 K.

parison in Figs. 2(b) and 6(b). To the best of our knowledge, there has been no prior reported theoretical value in the literature for the Seebeck coefficients of AlInN alloys. Future theoretical studies by using Boltzmann transport equation³⁷ or iterative method⁴⁶ are important to extract the Seebeck coefficients of the AlInN alloys, in order to provide directions for optimization of AlInN alloys as thermoelectric materials.

The findings presented here are important for providing useful thermoelectric parameters of III-Nitride and AlInN alloys in the literature.^{47–49} Future works include comprehensive measurements of thermal conductivities of these n-type AlInN alloys with various In-contents up to high temperature, in order to obtain the Z^*T values of these materials at high temperature operation. The measurements of the thermal conductivities of these alloys can be carried out by employing the 3ω measurement setup with differential^{50,51} and slope^{52–55} methods, similar to the approaches used in Refs. 38–42.

V. SUMMARY

In summary, the Seebeck coefficient characteristics of MOVPE-grown n-type $Al_{1-x}In_xN$ (x = 0.0038-0.2134) alloys are presented from T = 300 up to T = 382 K. The Seebeck coefficients were also characterized for InN and AlN binary

alloys for comparison purpose. The Seebeck coefficients show significant enhancement at higher temperatures for the n-type $Al_{1-x}In_xN$ alloys. In addition, the Seebeck coefficients for n-type n-Al_{1-x}In_xN alloy show decreasing trend with increasing In-content in the alloy. The Seebeck coefficients for the lattice-matched Al_{0.83}In_{0.17}N ($n = 5.1 \times 10^{18} \text{ cm}^{-3}$) sample were measured as high as 602.0–1233.2 μ V/K in the temperature range of T = 300 K up to T = 382 K, which are significantly higher in comparison with those measured for RF-sputtered AlInN (20–50 μ V/K, T = 300-400 K)²⁸ and MOVPE-grown InGaN (150–180 μ V/K, T = 300–400 K).^{38,39} The enhancement of the Seebeck coefficients can be attributed to longer relaxation time of carriers of the MOVPE-grown AlInN alloys with better crystalline quality. The increase of Seebeck coefficients of the n-type Al_{1-x}In_xN alloys at elevated temperature (T = 382 K) can lead to ~ 1.5 -4 times enhancement of the thermopower over those obtained at room temperature, which indicates that n-type AlInN materials as promising thermoelectric materials at high device operating temperature.

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