APPLIED PHYSICS LETTERS **92**, 042112 (2008)

Thermoelectric properties of $In_xGa_{1-x}N$ alloys

B. N. Pantha, R. Dahal, J. Li, J. Y. Lin, and H. X. Jiang[a)]
Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601, USA

G. Pomrenke
Directorate of Physics and Electronics, Air Force Office of Scientific Research, Arlington, Virginia 22203-1768, USA

(Received 13 December 2007; accepted 8 January 2008; published online 30 January 2008)

Thermoelectric (TE) properties of $In_xGa_{1-x}N$ alloys grown by metal organic chemical vapor deposition have been investigated. It was found that as indium concentration increases, the thermal conductivity decreases and power factor increases, which leads to an increase in the TE figure of merit (*ZT*). The value of *ZT* was found to be 0.08 at 300 K and reached 0.23 at 450 K for $In_{0.36}Ga_{0.64}N$ alloy, which is comparable to those of SiGe based alloys. The results indicate that InGaN alloys could be potentially important TE materials for many applications, especially for prolonged TE device operation at high temperatures, such as for recovery of waste heat from automobile, aircrafts, and power plants due to their superior physical properties, including the ability of operating at high temperature/high power conditions, high mechanical strength and stability, and radiation hardness. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2839309]

Thermoelectric (TE) devices convert heat energy directly into electrical energy without any moving parts. These devices are environmentally friendly because they produce no ozone-depleting gases and emit no radioactive radiation.[1] The best TE materials for room temperature application are Bi_2Te_3 based materials and structures,[2,3] but their applications are limited because tellurium is scarce, volatile, and toxic. Furthermore, the operational range of these materials is limited to temperatures lower than ~100 °C. Therefore, the search of materials for TE applications beyond the tellurium based compounds is necessary. Some of the outstanding features of III-nitrides that are highly attractive for TE applications include the ability for high power and high temperature operation, high mechanical strength and stability, and radiation hardness. Additionally, III-nitrides potentially offer tremendous scope for the enhancement of the TE figure of merit (*ZT*) via alloying, bandgap engineering and nanostructure incorporation with great ease. However, only a few reports concerning the thermal and TE properties of III-nitrides have been documented.[4–7] Here, we report on the experimental investigation of InGaN alloys as TE materials. It was observed that the thermal conductivity of $In_xGa_{1-x}N$ alloys substantially decreases with an increase of In-content. It was found that both the power factor and *ZT* systematically increase with an increase of the indium content. More importantly, *ZT* values of $In_{0.36}Ga_{0.64}N$ are comparable to those of SiGe alloys,[8] which is the current prime choice of TE materials for thermopower generation in high temperature environment. The initial results indicate that $In_xGa_{1-x}N$ alloys could be potentially very important TE materials.

The performance of TE materials is characterized by the TE figure of merit *ZT* ($=S^2\sigma T/\kappa$, where S=Seebeck coefficient, σ=electrical conductivity, κ=thermal conductivity, and T=absolute temperature). While there is no fundamental upper limit to *ZT*, progress has been extremely hard to come by, mainly due to the coupling between S, σ, and κ—changing one parameter would alter the others. There has been much attention drawn to finding TE materials suitable for solid-state refrigeration and power conversion. In particular, thin film TE materials are of great interest because they offer the potential for direct integration of microcoolers/power generators with various photonic and electronic devices.

Since the thermal conductivities of binary III-nitride crystals such as InN, GaN, and AlN are so high (>100 W/m K) (Refs. 9–11) that their prospects for TE device applications are limited. The current research in thin film TE materials without tellurium is concentrating on materials such as Si/Ge, SiGe/Si, and ErAs:InGaAs/InGaAlAs supperlattices and SiGe alloys.[12–16] It has been observed that alloying significantly reduces κ in solids with a very little deterioration of electrical properties[3–9,12–16] and making alloys a good candidate as TE materials. Here, we investigate the TE properties of $In_xGa_{1-x}N$ alloys grown by metal organic chemical vapor deposition.

$In_xGa_{1-x}N$ alloys (~110 nm in thickness) were grown on GaN/sapphire (0001) templates (reference sample). The indium concentrations were determined by $\theta/2\theta$ scans of the (0002) reflection using x-ray diffraction. Thermal conductivity and Seebeck coefficients measurements were performed by the differential 3ω and thermal gradient methods.[16–20] The material structures and heater/sensor geometry employed for the thermal conductivity measurements are schematically illustrated in Fig. 1(a). Since both $In_xGa_{1-x}N$ samples and GaN/sapphire templates (reference samples) are electrically conductive, we deposited 150 nm SiO_2 on the surfaces of both structures prior to the deposition of line heater/sensor. This layer provides the electrical insulation required for the 3ω measurements. Then, heaters/sensors of identical geometry, composed of Ni (20 nm)/Au (130 nm) with the wire width and length of 12 and 1000 μm, respectively, were patterned on the top of the SiO_2 layer of both structures using optical photolithography followed by metal deposition and lift-off techniques. A digital lock-in amplifier was used to feed the sinusoidal current into the specimen at an angular frequency ω and collect the voltage at a frequency 3ω across

[a)]Electronic mail: jiang@phys.ksu.edu.

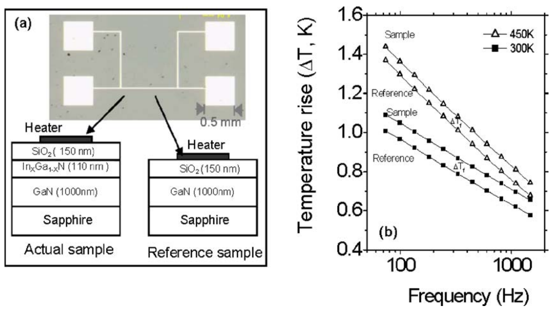

FIG. 1. (Color online) (a) Schematic illustration of cross sections and optical photograph of the top surface of a fabricated sample for 3ω measurements. (b) In-phase components of the temperature oscillation (ΔT) of the heater/sensor as a function of frequency of the driving current measured at 300 and 450 K for the reference (GaN/sapphire template) and an actual sample ($In_{0.28}Ga_{0.72}N$ epilayer grown on GaN/sapphire template).

the metal heater/sensor. Our detection system was calibrated by measuring κ values of sapphire and SiO_2 with measured κ values being 27.7 and 1.16 W/m K respectively, which agree well with literature values.[7,21,22]

Figure 1(b) shows the temperature oscillation ΔT in the line heater/sensor patterned on one of the $In_xGa_{1-x}N$ thin films (an actual sample) and its reference sample (GaN/sapphire template) as a function of frequency of the driving current measured at 300 and 450 K. The temperature drop across the film ΔT_f was calculated by subtracting ΔT of a reference sample from ΔT of the actual sample. The mean value of ΔT_f in the entire frequency range was used to calculate κ. Standard deviations in measured ΔT_f were less than 3%. At 300 K, the measured κ of $In_xGa_{1-x}N$ alloys are 8.1, 5.4, 2.7, and 1.05 W/m K for x=0.16, 0.22, 0.28, and 0.36, respectively. Figure 2 shows the variation of κ with x, for $In_xGa_{1-x}N$ alloys measured at 300 K. Results for $Al_xGa_{1-x}N$ (Ref. 4) and $In_xAl_{1-x}N$ (Ref. 5) alloys are also included for comparison. A significant reduction in κ of $In_xGa_{1-x}N$ alloys with increasing x is observed, which is mainly attributed to the scattering of phonons due to the alloy disorder.[7,9,23,24] The κ values of $In_xGa_{1-x}N$ alloys are comparable to those of other nitride systems, such as $In_xAl_{1-x}N$ and $Al_xGa_{1-x}N$.[4–6]

In order to evaluate the TE figure of merit (ZT), Seebeck coefficients (S), and electrical conductivities (σ) of $In_xGa_{1-x}N$ alloys were measured by a temperature gradient method[16] and van der Pauw Hall-effect measurement[25] in the in-plane direction. Figure 3(a) shows the measured S and σ of $In_xGa_{1-x}N$ alloys as functions of In content ($0<x<0.36$). S decreases while σ increases with an increase of x. The reason for this trade-off relationship between S and σ is due to the increased background electron concentration with an increase of x in $In_xGa_{1-x}N$ alloys. In Fig. 3(b), we plot the power factor ($P=S^2\sigma$) and ZT as functions of In content. Both P and ZT are observed to increase with x. At 300 K, ZT=0.08 is obtained in the $In_{0.36}Ga_{0.72}N$ alloy, which is much larger than those in AlInN (Ref. 5) and AlGaN (Ref. 7) alloys in which ZT is around (or even smaller than) 0.001 at 300 K. This may be due to the fact that phonons are more easily scattered in $In_xGa_{1-x}N$ alloys than in the other two ternary alloy systems, attributing to a more pronounced local strain-scattering effect because In atoms are larger than Ga and Al.[24] Furthermore, for materials with comparable free carrier concentrations, InGaN has a higher electron mobility than AlGaN or AlInN. The issue of high background electron concentration in InGaN alloys with relatively high In contents is currently under intensive investigation[26,27] and significant improvements in InGaN material quality and conductivity control are anticipated, which will lead to further enhancement in ZT and P in InGaN alloys with larger In contents.

The temperature dependent TE properties of $In_xGa_{1-x}N$ alloys were also measured in the temperature range from 300 to 450 K. Figure 4 shows ZT of $In_{0.36}Ga_{0.72}N$ alloy as a function of temperature. Data for SiGe alloys[8] are also in-

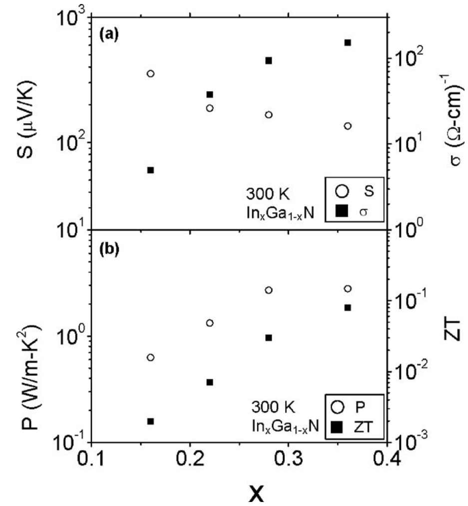

FIG. 3. (a) Seebeck coefficient (S) and electrical conductivity (σ) of $In_xGa_{1-x}N$ alloys as functions of In content (x). (b) Power factor ($P=S^2\sigma$) and figure of merit (ZT) of $In_xGa_{1-x}N$ alloys as functions of In content (x) measured at 300 K.

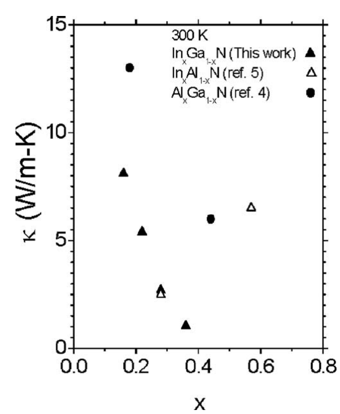

FIG. 2. Thermal conductivity (κ) of $In_xGa_{1-x}N$ alloys as a function of In content (x) at 300 K. Data points for AlGaN and InAlN (from Refs. 4 and 5) are included for comparison.

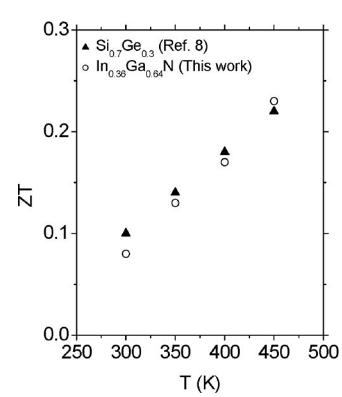

FIG. 4. Measured ZT values of $In_{0.36}Ga_{0.64}N$ alloy from 300 to 450 K. Data for SiGe alloys from Ref. 8 are included for comparison.

cluded for comparison. The *ZT* values of $In_{0.36}Ga_{0.64}N$ alloy in the measured temperature range are comparable to those of the SiGe alloys, which is the current prime choice of TE materials for thermopower generation in high temperature environment such as in radio-isotope thermoelectric generators. We observe that *ZT* increases linearly with temperature and reaches a value of 0.23 at 450 K. The results suggest that $In_xGa_{1-x}N$ alloys may replace SiGe alloys for applications of prolonged TE device operation at high temperatures, such as for recovery of waste heat from automobile, aircrafts, and power plants due to their superior physical properties over those of SiGe, including the ability of operating at high temperature/high power conditions and high mechanical strength and stability.

Significant enhancement in the TE figure of merit of InGaN is anticipated due to the fact that III-nitrides potentially offer tremendous scope for the enhancement of *ZT* values via the bandgap engineering and nanoscale structuring, which were shown in other material systems to allow one to either use quantum confinement of carriers or spectrally dependent scattering of phonons to manipulate S, σ, and κ in ways that can increase *ZT* beyond the bulk values.[3,28] Furthermore, III-nitride planar processing is now well developed for photonic/electronic devices and sensors, which can facilitate relatively low cost fabrication by the use of existing semiconductor processing equipment. Finally, future communication platforms will employ highly sophisticated micro-nanoscale sensor systems in which III-nitride based devices will play increasingly important roles because of their superior material properties and the development of III-nitride based TE devices may open up the possibility for monolithic integration of TE power generator/cooler modules onto remote micro-nanoscale sensor networks.

This work was funded in part by the U.S. Air Force Office for Scientific Research (Grant No. FA9550-06-1-0441) and Department of Energy (Award No. 96ER45604).

[1]G. Mahan, B. Sales, and J. Sharp, Phys. Today **50**, 42 (1997).
[2]M. G. Kanatzidis, S. D. Mahanti, and T. P. Hogan, *Chemistry, Physics, and Materials of Science and Thermoelectric Materials* (Kluwer Academic/Plenum, New York, 2002).
[3]R. Venkatasubramanian, E. Slivola, T. Colpitts, and B. O'Quinn, Nature (London) **413**, 597 (2001).
[4]B. C. Daly, H. J. Maris, A. V. Nurmikko, M. Kuball, and J. Han, J. Appl. Phys. **92**, 3820 (2002).
[5]S. Yamaguchi, R. Izaki, K. Yamagiwa, K. Taki, Y. Iwamura, and A. Yamamoto, Appl. Phys. Lett. **83**, 5398 (2003).
[6]S. Yamaguchi, Y. Iwamura, and A. Yamamoto, Appl. Phys. Lett. **82**, 2065 (2003).
[7]W. Liu and A. A. Balandin, J. Appl. Phys. **97**, 123705 (2005).
[8]J. P. Dismukes, L. Ekstrom, E. F. Steigmeier, I. Kudman, and D. S. Beers, J. Appl. Phys. **35**, 2899 (1964).
[9]J. Piprek, *Semiconductor Optoelectronic Devices* (Academic, San Diego, 2003).
[10]K. E. Sichel and J. I. Pankove, J. Phys. Chem. Solids **38**, 330 (1977).
[11]G. A. Slack, R. A. Tanzilli, R. O. Pohl, and J. W. Vandersande, J. Phys. Chem. Solids **48**, 641 (1987).
[12]S. M. Lee, D. G. Cahill, and R. Venkatasubramanian, Appl. Phys. Lett. **70**, 2957 (1997).
[13]S. T. Huxtable, A. R. Abramson, C. L. Tien, A. Majumdar, C. LaBounty, X. Fan, G. Zeng, J. E. Bowers, and A. Shakouri, Appl. Phys. Lett. **80**, 1737 (2001).
[14]G. Zeng, J. E. Bowers, J. M. O. Zide, A. C. Gossard, W. Kim, S. Singer, A. Majumdar, R. Singh, Z. Bian, Y. Zhang, and A. Shakouri, Appl. Phys. Lett. **88**, 113502 (2006).
[15]M. Takashiri, T. Borca-Tasciuc, A. Jacquot, K. Miyazaki, and G. Chen, J. Appl. Phys. **100**, 054315 (2006).
[16]B. Yang, W. L. Liu, J. L. Liu, K. L. Wang, and G. Chen, Appl. Phys. Lett. **81**, 3588 (2002).
[17]D. G. Cahill, Rev. Sci. Instrum. **61**, 802 (1990).
[18]D. G. Cahill, M. Katiyar, and J. R. Ablson, Phys. Rev. B **50**, 6077 (1994).
[19]L. Lu, W. Yi, and D. L. Zhang, Rev. Sci. Instrum. **72**, 2996 (2001).
[20]C. Dames and G. Chen, Rev. Sci. Instrum. **76**, 124902 (2006).
[21]S. M. Lee and D. G. Cahill, J. Appl. Phys. **81**, 2590 (1997).
[22]K. A. McCarthy and S. S. Ballard, J. Opt. Soc. Am. **41**, 1062 (1951).
[23]S. Adachi, J. Appl. Phys. **54**, 1844 (1983).
[24]G. S. Nolas, J. Sharp, and H. J. Goldsmid, *Thermoelectrics Basic Principles and New Materials Development*, Springer Series in Material Science Vol. 45 (Springer, New York, 2001), pp. 79–82.
[25]L. J. van der Pauw, Philips Res. Rep. **13**, 1 (1958).
[26]C. G. Van de Walle and D. Segev, J. Appl. Phys. **101**, 081704 (2007).
[27]P. D. C. King, T. D. Veal, C. F. McConville, F. Fuchs, J. Furthmüller, F. Bechstedt, P. Schley, R. Goldhahn, J. Schörmann, D. J. As, K. Lischka, D. Muto, H. Naoi, Y. Nanishi, H. Lu, and W. J. Schaff, Appl. Phys. Lett. **91**, 092101 (2007).
[28]L. D. Hicks, T. C. Harman, X. Sun, and M. S. Dresselhaus, Phys. Rev. B **53**, R10493 (1996).