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ScienceDirect
Thin Solid Films 515 (2007) 4501 – 4504
www.elsevier.com/locate/tsf

Thermoelectric properties and thermoelectric devices of free-standing GaN and epitaxial GaN layer

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Available online 8 December 2006

Abstract

We studied the thermoelectric properties of free-standing GaN (fr-GaN) and epitaxial GaN layer (epi-GaN), and furthermore, we have fabricated thermoelectric devices using these materials. For fr-GaN, the maximum power factor was 7.7×10^{-4} W/m K^2 at 373 K, and for epi-GaN layer, the maximum power factor was 9.4×10^{-4} W/m K^2 at 373 K. The devices fabricated are (a) fr-GaN and chromel of 4 pairs, and (b) epi-GaN and chromel of 3 pairs. The maximum output power and the open output voltage were (a) 3.35×10^{-6} W and 2.76×10^{-2} V at ΔT=153 K, and (b) 1.21×10^{-7} W and 1.71×10^{-2} V at ΔT=153 K, respectively.

Keywords: GaN; HVPE; Thermoelectric

1. Introduction

Thermoelectric conversion that can convert the thermal energy directly into electrical energy has lately attracted considerable attention as environmental and energy-saving technology which can recycle the waste heat energy. However, thermoelectric conversion has been limited to commercial use because the highest performing thermoelectric materials which are intermetallic compounds such as Bi_2Te_3 and PbTe have poor thermal durability, high toxicity, and high cost. Accordingly, recent research has focused on new thermoelectric materials which are several environmentally conscious materials such as $NaCo_2O_4$ of oxide compounds.

Currently, as is typical of ubiquitous computing network, the demand of downsizing electric devices is increasing. Low-power electric device will become more important now. Thermoelectric conversion is expected for application to various fields, such as bio, medical devices, and wearable computing, such as an independent power supply for low electric power devices.

III-nitride semiconductors have high prospects for applications in optical and electronic devices, and already been commercialized, such as light-emitting devices by process technology at a nano-scale. III-nitrides have the advanced processes necessary for achieving thermoelectric devices such as independent power supplies for low electric power devices. Moreover, III-nitrides have many advantages, as compared with Bi_2Te_3 system, which have low environmental stress, low cost, high-melting point, mechanical and chemical stability, and more.

We have earlier studied the thermoelectric properties of amorphous AlInN and InN thin films prepared by reactive radio-frequency (RF) sputtering method [1–6]. However, the physical properties of amorphous nitrides are not understood at present. In this study, we have investigated the thermoelectric properties of single-crystalline free-standing GaN (fr-GaN) and epitaxial GaN layers (epi-GaN). Since physical properties of single-crystalline nitrides were already clarified, material design and device design can be facilitated by using these materials in practical use. As the demonstration of the material design, we calculated the electric resistivity and the Seebeck coefficient of

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0040-6090/$ - see front matter © 2006 Published by Elsevier B.V.
doi:10.1016/j.tsf.2006.07.145

Table 1
Parameters used in the calculation

ε	9.4
ε_∞	5.2
E_1 [eV]	8.4
C_1 [N/cm^2]	2.65×10^7
m_e^*	0.19
ρ [g/cm^3]	6.07
T_{po} [K]	1000
N_D [cm^{-3}]	1.5×10^{18}
N_A^{net}/N_D	0.35

fr-GaN, and compared it with the experiment value. Furthermore, we have fabricated thermoelectric devices using these materials.

2. Experimental procedure

Fr-GaN and epi-GaN were prepared by HVPE method [7–9]. Epi-GaN was deposited on sapphire substrate. Si was doped into those materials as n-type dopant. The thickness of fr-GaN was 390 μm, and the thickness of epi-GaN was 35 μm. For fr-GaN, the carrier concentration and mobility were about 1×10^{18} cm^{-3} and 250 cm^2/Vs at room temperature, and for epi-GaN layer, the carrier concentration and mobility were about 6×10^{18} cm^{-3} and 130 cm^2/Vs at room temperature, respectively. Their electrical resistivity (ρ) and Seebeck coefficients (α? were simultaneously measured by the dc method and the conventional four-probe method, respectively. Measurements were carried out in the temperature range 373 K to 973 K in low pressure He gas. To evaluate the potential of those materials as one of thermoelectric materials, we used the standard of power factor ($=\alpha^2/\rho$).

To compare experimental values with calculation values, we calculated ρ and α of fr-GaN. These were calculated under degenerate conditions.

$$n = N_c F_{\frac{1}{2}}(\zeta) \tag{1}$$

Here, n is the electron carrier concentration, N_c is the effective density of states of the conduction band, and $F_{1/2}(\zeta)$ is the Fermi integral shown below. ζ is the reduced Fermi energy, E_F/k_BT (4).

$$F_j(\zeta) = \frac{1}{j!}\int_0^\infty \frac{\xi^j \mathrm{d}\xi}{\exp(\xi-\zeta)+1} \tag{2}$$

The electron mobility μ in GaN is obtained from $\mu^{-1} = \mu_i^{-1}+\mu_{ac}^{-1}+\mu_{po}^{-1}$, where μ_i^{-1}, μ_{ac}^{-1}, and μ_{po}^{-1} originate from the scattering by ionized impurities, acoustic phonons, and polar optical phonons, respectively.

$$\mu_i = \frac{2^{\frac{15}{2}}\pi^{\frac{1}{2}}\varepsilon^2(k_BT)^{\frac{3}{2}}}{e^3 n_i m^{*\frac{1}{2}}} G(T,n) \tag{3}$$

$$\mu_{ac} = \frac{2^{\frac{3}{2}}\pi^{\frac{1}{2}} e\hbar^4 C_1}{3m^{*\frac{1}{2}} E_1^2 (k_BT)^{\frac{3}{2}}} \tag{4}$$

$$\mu_{po} = \frac{2^{\frac{9}{2}}\pi^{\frac{1}{2}}}{3}\frac{\hbar^2(k_BT)^2(\exp(T_{po}/T)-1)\chi(T_{po}/T)}{e(k_BT_{po})m^{*\frac{3}{2}}(\varepsilon_\infty^{-1}-\varepsilon^{-1})} \tag{5}$$

Here, $G(T, n)$ is a function of T and n (4) and $\chi(T_{po}/T)$ is a function of T_{po}/T (5). In the n-type sample and the donor-dominant case, the charge balance equation is given as

$$n + N_A^{net} = N_D^+(= N_D - n_D) \tag{6}$$

where the compensation rate N_A^{net}/N_D and N_D are fitting parameters, and E_D, which is a parameter of n_D, is −30 meV (6). In the Fermi statistics system, the Seebeck coefficient is obtained by the following expression (7).

$$\alpha = -\frac{k_B}{e}\left(\frac{(r+5/2)F_{r+3/2}}{(r+2/3)F_{r+1/2}} - \zeta\right) \tag{7}$$

Other notations not referred to have the standard meanings as shown in Table 1. Using Eqs. (1)–(7), we calculated the temperature dependence of ρ $(=(en\mu)-1)$ and α.

We fabricated thermoelectric devices using fr-GaN and epi-GaN. The devices are composed of (a) fr-GaN and chromel of 4 pairs, and (b) epi-GaN and chromel of 3 pairs on SiO_2 glass substrate. Chromel was used instead of p-type nitrides because the sign of its Seebeck coefficient is positive. Applying the temperature difference (ΔT) between the edges of device, the output power and the output voltage, namely, the load characteristic, were measured. One side of the device was heated to a higher temperature (T_h) by heating the block with voltage, while the low temperature (T_c) side was kept at room temperature. Thus, a temperature difference ΔT $(=T_h-T_c)$ was applied between the two sides.

3. Result and discussion

Fig. 1 shows temperature dependence of the ρ of fr-GaN in the y-axis. The closed circles and crosses represent experiment values and calculation values, respectively. The ρ values increased together with increasing temperature. This is typical electrical behavior of metallic or degenerately doped semiconductors with high carrier concentration. This result shows some discrepancies between the experiment values and the calculation ones at high temperatures. In this calculation, the scattered mechanism assumed only the phonon scattering ($r=-1/2$).

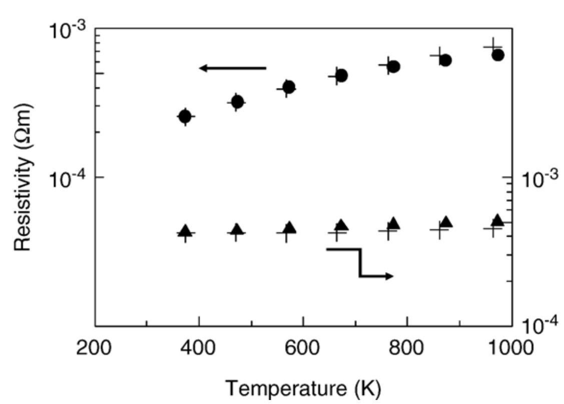

Fig. 1. Temperature dependence of electrical resistivity (left ordinate) and Seebeck coefficient (right ordinate) of fr-GaN of experimental values (closed circles) and calculation values (crosses).