Thermal conductivity of bulk GaN single crystals

A. Jeżowski, [a,1] P. Stachowiak, [a] T. Suski, [b] S. Krukowski, [b] M. Boćkowski, [b] I. Grzegory, [b] and B. Danilchenko [c]

[a] *Institute of Low Temperature and Structure Research, P.O. Box 1410, 50-950 Wrocław, Poland*
[b] *High Pressure Research Center, Sokołowska 29/37, 01-142 Warsaw, Poland*
[c] *Institute of Physics, Prosp. Nauki 46, Kiev, Ukraine*

Abstract

We have measured thermal conductivity, κ, in the wide temperature range 4–300 K of GaN bulk single crystals grown by high–pressure, high–temperature synthesis. Obtained results (1600 W/Km at 45 K) are the highest κ values reported on GaN material. At the room temperature κ is about 210 W/Km. The contributions to the GaN thermal resistance of Umklapp process, mass point defects as well as phonon scattering on dislocations and sample boundary are discussed.

Key words: gallium nitride; thermal conductivity

Group–III nitrides (e.g. GaN) have attracted a large amount of attention for their application in high–power optoelectronic and electronic devices. Heat dissipation in these high–power and high–temperature applications has become an important design aspect. Investigation of thermal conductivity and heat dissipation mechanisms in GaN is important from both the application and fundamental science point of view.

The aim of this work is to determine temperature dependence of κ in GaN bulk monocrystals grown by high–pressure high–temperature synthesis. Actually, reliable data on κ versus temperature in GaN are not available. The presented hitherto data were obtained mainly in room temperatures, for samples fabricated on different substrates (see [1] and [2]). High pressure grown GaN [3] has been used as a substrate for high quality epitaxial layers, heterostructures and blue laser diode [4]. These GaN crystals are characterized by n–type conductivity originating from unintentionally introduced oxygen donors. Dislocation concentration is below 10^2 cm^{-2}. Since the low structural quality and built–in strain present in the epitaxial GaN layers grown on foreign substrates, high pressure grown GaN crystals are often employed as a bunch material for determination of basic physical properties of GaN.

Three samples of bulk GaN, obtained at various conditions, were used in the present experiment. They were characterized by X-ray diffraction methods and electrical measurements. Full Width at Half Maximum for the X-ray peak of (0004) reflection gave the value between 25 and 40″ arc showing high monocrystallinity of the material. Since no evidence of low angle boundaries were evidenced, one can conclude that the possible contribution to thermal conductivity κ, originating from grain boundaries can be neglected. Hall effect measurements showed that the samples used are highly conductive and are characterized by electron concentration of about 5×10^{19} cm^{-3}.

Thermal conductivity measurements were carried out by the axial stationary heat flow method in the temperature range 4–300 K on a parallelepiped samples ($5 \times 3 \times 0.04$ mm).

The results of measurements of their thermal conductivity dependence on temperature are presented in Fig. 1. The common feature of the curves is starting with the increase of κ at low temperatures. Next, the thermal conductivity coefficients reach maximum values in the range 45–55 K and the fall down at further in-

[1] Corresponding author. E-mail: an_je@int.pan.wroc.pl

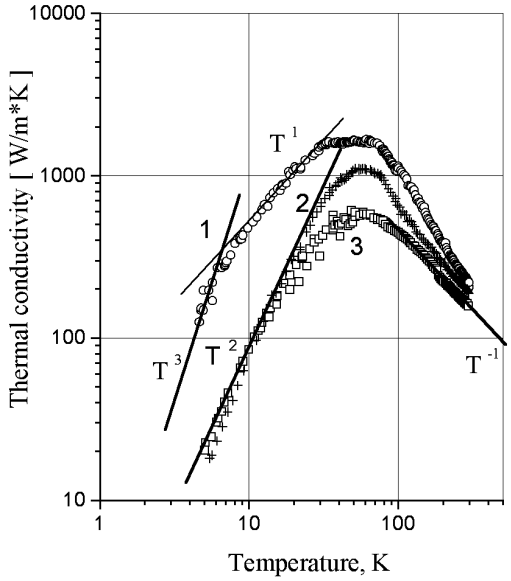

Fig. 1. The temperature dependence of κ of the bulk GaN samples grown in different conditions. Straight lines show the asymptotic behavior in the different temperature ranges.

crease of temperature. The sample 1 exhibits the highest conductivity at the maximum; its character of $T(\kappa)$ dependence is different from that of the samples 2 and 3. At the lowest temperature, the sample 1 shows behavior asymptotic to T^3. Such a behavior is typical for perfect crystals in which the phonon free path becomes equal the sample dimensions. At somewhat higher temperatures the dependence becomes weaker. The character of $T(\kappa)$ changes at these temperatures may be connected with an increase of phonon scattering in processes for which relaxation time rises rapidly with phonon frequency (like, e.g. scattering on point defects – oxygen atoms). On the other hand, the samples 2 and 3 exhibit another dependence on temperature, which is closer to T^2. This dependence can be attributed to phonon scattering on the extended defects of structure. It is reasonable to associate this kind of structure imperfections with planar defects and a small amount of dislocations.

At temperatures above the maximum, the thermal conductivity exhibits different picture of dependence. The crystal 3 shows conductivty almost inversely proportional to the temperature. The samples 1 and 2 behavior deviates from this simple law. It indicates that the thermal resistance at high temperatures of such GaN samples could deal with Umklapp process of phonon scattering. Indeed, involving this processes allow us to describe the temperature dependence of thermal resistance (ω) in the simplest way.

In the approximation of the most relevant phonons [5], the thermal resistance can be approximated with the following expression:

$$\omega = \left\{ \frac{0.25}{T^3} + \frac{1.7\times10^{-2}}{T} + 4.5\times10^{-6}T + \right.$$
$$\left. +5.5\times10^{-3}\left(\frac{\Theta_D}{3T}\right)^{-1}\exp\left[-\frac{\Theta_D}{3T}\right]\right\}\,[\mathrm{K/Wm}]\,.$$

The first term in this expression deals with phonon scattering on the sample boundary. The second one can be associated with the phonon scattering by isolated dislocations. The third term is related to mass point defects scattering and the last term describes the non–elastic phonon scattering in the Umklapp process. Here Θ_D is the Debye temperature for GaN.

The thermal resistance ($\omega = 1/\kappa$) dependence on temperature for the sample 1 fits well to the above expression. The best fit of experimental data for the sample 1 was obtained for $\Theta_D = 690\,\mathrm{K}$.

Concluding, undoped crystals of GaN exhibit very high thermal conductivity which value (mainly at the maximum) and temperature dependence at low temperatures is strongly connected with the defect level in a sample. Point defects and dislocations exert substantial influence on reducing the thermal conductivity. At room temperature, where the phonon–phonon scattering dominates, the influence of defects and dislocations is weaker, which is evidenced in smaller variation of κ (160 W/Km – sample 3; 210 W/Km – sample 1). The quantitative analysis of the observed phenomena calls for further investigation. Its results will be published elsewhere.

Acknowledgements

This work was supported by the Polish State Committee for Scientific Research (Grant No. 7 T08A 008 21) and USF (Grant No. F7/379).

References

[1] D. I. Florescu, V. M. Asnin, F. H. Pollak, A. M. Jones, J. C. Ramer, M. J. Schurman, and I. Ferguson, Appl. Phys. Lett. **77** (2000) 1464.

[2] D. I. Florescu, V. M. Asnin, and F. H. Pollak, Compound Semicond. **7** (2001) 62.

[3] S. Porowski. J. Cryst. Growth **189–190** (1998) 153.

[4] I. Grzegory, S. Krukowski, M. Leszczyński, P. Perlin, T. Suski, and S. Porowski, Acta Phys. Polon. **100** (Suppl.) (2001) 57.

[5] R. Berman, Thermal conduction in solids (Clarendon, Oxford 1976).