Pis'ma v ZhETF, vol. 112, iss. 2, pp. 112 – 113

High thermal conductivity of bulk GaN single crystal: An accurate experimental determination

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Submitted 19 May 2020
Resubmitted 1 June 2020
Accepted 2 June 2020

DOI: 10.31857/S1234567820140086

In recent decades, GaN crystals attract a lot of attention, both experimental and theoretical, due to its unique physical properties, including thermal conductivity. In pure and lightly doped GaN, the heat is transported practically solely by phonons.

Experiments demonstrate that defects may reduce strongly the thermal conductivity of GaN crystals in a wide temperature range [1–8]. Oxygen substitutional atoms, gallium vacancies, and their complexes are the most common point defects in unintentionally doped GaN. The oxygen atoms at concentration above $10^{18}\,\mathrm{cm}^{-3}$ produce substantial decrease in κ at room temperature, and in heavily oxygen doped GaN ($> 10^{19}\,\mathrm{cm}^{-3}$) an additional scattering process owing to the phonon interaction with free electrons supplied by oxygen donors becomes important [5].

The impact of point defects on thermal conductivity of GaN crystals was investigated by employing the phenomenological models [1, 3, 5–9] and first-principles approaches [10, 11]. For the GaN crystal studied in [1], the contribution of point-defect scattering toward thermal resistivity was estimated in [9] to be as high as 90 % around the peak of $\kappa(T)$ at $\sim 30\,\mathrm{K}$ and about 11 % at 300 K. Lindsay et al. have shown by *ab-initio* calculations [12, 13] that the strong effect of impurities is due to small contribution of anharmonic phonon-phonon scattering processes to the phonon relaxation. This is a consequence of large gap between acoustic and optic modes' frequencies in the phonon spectra of GaN, which arises due to the high mass ratio of Ga and N atoms.

To clarify the effect of defects in the heat transport of GaN we performed an accurate measurements of GaN single crystal over a wide temperature range.

The wurtzite GaN single crystal plate was grown by hydride vapor-phase epitaxy (HVPE) on a sapphire substrate. After the growth upon cooling, the plate self-separated from the substrate [14]. The bar-shaped sample with the cross-section of $1.38 \times 3.24\,\mathrm{mm}^2$ and the length of 6.5 mm was made from the plate. We employed a steady-state longitudinal heat flow technique for thermal conductivity measurements within the basal plane (see [15] for details).

From the measured Raman spectra, the free electron concentration n_e was estimated to be $(2.7 \pm 0.2) \times \times 10^{17}\,\mathrm{cm}^{-3}$ for our sample. This value is very close to the concentration of spin 1/2 paramagnetic defects, $(2{-}3) \times 10^{17}\,\mathrm{cm}^{-3}$, that was determined by DC magnetization measurements using SQUID magnetometer. We suppose that impurity oxygen and silicon, the shallow donors in GaN, are responsible for the paramagnetism in our GaN crystal at low temperature when being in the neutral charge state, whereas they supply with free charge carriers at room temperature.

The measured $k(T)$ for our GaN crystal is presented in Fig. 1. Here, plotted are the experimental data for bulk GaN of other works [1, 2, 4, 5, 16]. In general, there is a good agreement between results of different measurements. Some essential discrepancies, however, emerge at close inspection. We have found that $\kappa(T) \propto T^{-n}$ with $n = 1.367 \pm 0.002$ in the range $80 < T < 300\,\mathrm{K}$, but Slack et al. have observed a weaker dependence $\kappa(T) \propto T^{-1.22}$. According to the experimental results of [7] the slope decreases with increasing doping from $n = 1.3$ for the undoped sample to $n = 0.55$ for the highest Si-doped sample.

At lowest temperatures of our experiment, below about 5.5 K, the measured values of $\kappa(T)$ are close to the calculated ones, shown as the violet line in Fig. 1. The calculations were performed assuming only the diffuse scattering from sample boundaries and taking into account the phonon focusing effect. The measured $\kappa(T)$ at 5 K is about 13 % lower than the calculated value.

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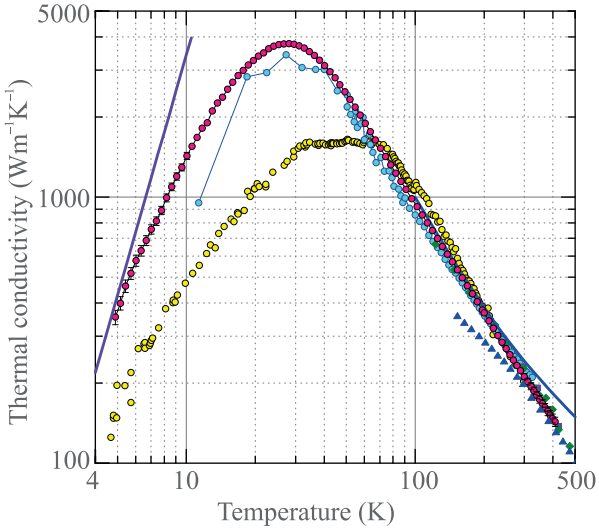

Fig. 1. (Color online) Thermal conductivity of GaN single crystals as a function of temperature. Pink circles indicate measurement data of this work, the violet line is a calculated T^3-dependence in the diffuse boundary scattering regime. The published results of other experiments – Slack et al. [1] (cyan circles), Jeżowski et al. [2] (yellow circles), Mion et al. [4] (violet squares), Simon et al. [5] (olive diamonds), and Zheng et al. [16] (blue triangles) – and calculations by Lindsay et al. [12] (the solid blue line) are also shown

This suggests that even at this temperature the point defect scattering sizably reduces the conductivity in our sample. The experimental curve $\kappa(T)$ progressively deviates from the calculated one with the temperature rise reflecting the increasing contribution of the point defect scattering as compared with boundary scattering strength. The $\kappa(T)$ for our GaN sample reaches the maximum of 3770 W m^{-1} K^{-1} at 28 K. Here, the value of $\kappa(T)$ is determined by the combined effect of the boundary, point-defect, and anharmonic scattering.

There is a very weak dip in the $\kappa(T)$ curve centered near $T \approx 7$ K (see Fig. 1). This dip can be ascribed to the effect of the phonon scattering from electrons bound to the neutral donors.

Comparing our measured $\kappa(T)$ with the calculated dependence of Lindsay et al. [12] for pure GaN (the blue solid line in Fig. 1), we find a very good quantitative agreement at temperatures from 100 to 200 K: the calculated values are higher than experimental ones by 4–5 %. This suggests the overwhelming dominance of three-phonon processes in the phonon scattering. The phonon scattering from the lattice imperfections and charge carriers both contribute small at these and higher temperatures. The rising deviation upward of the theoretical $\kappa(T)$ from experimental one with temperature increase above 200 K likely indicates the rising importance of four-phonon scattering processes in thermal conductivity.

Thermal conductivity, Raman, and magnetic measurements were carried out in the Resource Center funded by National Research Center "Kurchatov Institute". The authors are thankful to D. R. Streltsov for Raman measurements. A. V. Inyushkin, A. N. Taldenkov, and D. A. Chernodubov acknowledge financial support from the Russian Foundation for Basic Research (Grant # 19-07-00229).

Full text of the paper is published in JETP Letters journal. DOI: 10.1134/S0021364020140039

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