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Study of the Thermoelastic Effect in GaN

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Abstract. Young's modulus and internal friction of bulk GaN single crystal samples were measured as a function of temperature in 100 K to 700 K range. The internal friction strongly depends on sample dimensions and measurement frequency (3 – 10 kHz). The experimental values are compared to the theoretical model for thermoelastic damping. The model and the experimental data match very well in the whole temperature range and for different frequencies indicating that the thermoelastic damping is the main energy dissipation mechanism.

Introduction

Mechanical spectroscopy is a non-destructive technique that allows the measurement of the dynamic modulus and the internal friction. This last parameter accounts for the dissipation of mechanical energy in form of heat. In crystalline materials the main intrinsic energy loss mechanisms are: 1) thermoelastic damping caused by the strain-induced temperature gradients and 2) relaxations related to the mobility of crystal defects. The control of the presence of crystal defects and in particular dislocations is considered essential for the future applications of GaN which has gained a lot of attention in the last decades as a material of choice for electronic and opto-electronic devices. In fact, dislocations are a source of efficiency losses in GaN based LED since they act as non-radiative carrier recombination centers. The information about GaN mechanical properties is also of great importance for an effective application design. In this study, the measurements of Young's modulus and internal friction of a GaN bulk single crystal were made as a function of temperature for the first time. The measured internal friction is compared to the thermoelastic damping calculated using a theoretical approach.

Thermoelastic Effect

The theory of the thermoelastic damping due to the flow of transversal thermal current was first developed by Zener [1]. A very convincing experimental support of this theory was given soon after by Berry [2] who measured internal friction in brass samples at room temperature for different frequencies. The internal friction Q^{-1} depends on the oscillation frequency f, sample thickness a and several thermodynamic properties of the material. It can be modelled by the following equation:

$$Q^{-1} = \frac{\alpha^2 ET}{\rho c}\left(\frac{2\pi f\tau}{1+(2\pi f\tau)^2}\right) = \Delta\left(\frac{2\pi f\tau}{1+(2\pi f\tau)^2}\right), \quad (1)$$

where α is the thermal expansion coefficient, E the Young's modulus, T the temperature, c the specific heat, ρ the density and $\Delta = \frac{\alpha^2 ET}{\rho c}$ is the relaxation strength. The relaxation time τ is given by

$$\tau = a^2\rho c/\pi^2\lambda\,, \quad (2)$$

where λ is the thermal conductivity and a the thickness of the sample.

Experimental

The samples of dimensions 0.3 x 3 x 20 mm^3 were laser cut from an n-type GaN single crystal wafer. The wafer was grown by hydride vapour phase epitaxy under atmospheric pressure using H_2 as the carrier gas. The obtained single crystal had c-axis oriented wurtzite crystal structure, as confirmed by X-ray diffraction.

The mechanical spectroscopy measurements were made in a free-free vibrating reed installation [3]. The capacitive excitation was possible due to the rather high electrical conductivity of the sample. The Young's modulus E is calculated from the resonant frequency f of the first mode of flexural vibrations according to:

$$E = 0.9464 \cdot \rho \cdot \frac{l^4}{a^2} \cdot f^2, \tag{3}$$

where l is the sample length. The density calculated from the sample mass and dimensions is found to be $\rho = (6.00 \pm 0.05)$ g/cm^3. The internal friction Q^{-1} was obtained from an exponential fit of the free decay of the oscillations:

$$A(t) = A_0 e^{-Q^{-1}\pi f t}, \tag{4}$$

where A is the oscillation amplitude as a function of time t. Maximum oscillation strain was of the order of 10^{-5} and the heating and cooling rate 1 K/min or less under vacuum better than 10^{-4} mbar.

Measurement Results

The internal friction and resonant frequency measured in cooling from 700 to 100 K are presented in Fig. 1a. The internal friction remains almost flat down to 350 K, below which it strongly decreases. In parallel, the resonant frequency monotonously increases. The measurements on first heating are not presented as they exhibited some anomalies due to the hydrocarbon residues that disappear when the sample is heated above the cracking temperature (~ 600 K in our case). Further thermal cycles are overlapping the first cooling measurements showing very good reproducibility of this behaviour.

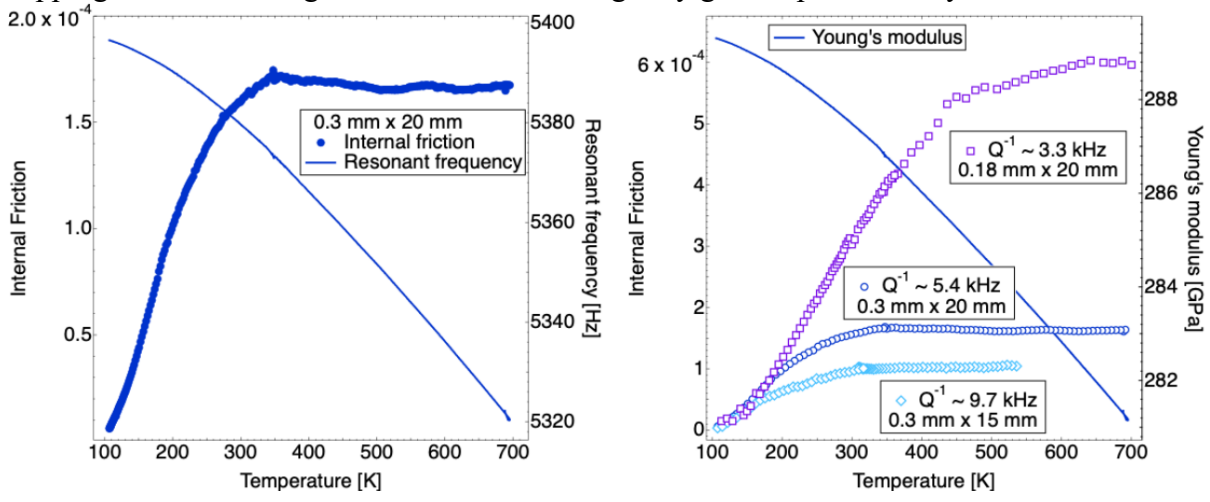


Fig. 1. a) Internal friction and resonant frequency of GaN single crystal as a function of temperature, measured on cooling for a sample of 0.3 mm thickness and 20 mm length. b) Temperature dependence of Young's modulus and internal friction for different sample dimensions and resonant frequencies. For clarity, only every 50-th measurement point is presented and the error bars on modulus of $\pm$ 10 GPa are not included.

By changing the sample dimensions, it is possible to change the resonant frequency according to Eq. 3. By reducing the sample length from 20 to 15 mm, its resonant frequency increases from around 5.4 kHz to 9.7 kHz. By keeping the original length of 20 mm and reducing the sample thickness by polishing, from 0.3 mm to 0.18 mm, the resonant frequency decreases to about 3.3 kHz. Fig. 1b shows how the measured internal friction is affected. For shorter sample oscillating at higher resonant frequency, the internal friction in the high temperature range is reduced and the temperature below which the internal frequency decreases is also slightly lower, about 320 K. For thinner sample and thus lower resonant frequency, the internal friction at high temperatures is much higher. Its decrease on cooling starts also at much higher temperature, around 650 K. The temperature dependence of the Young's modulus, calculated using the Eq. 3 and the resonant frequency presented in Fig. 1a, is also shown in Fig. 1b.

Theoretical Calculation

The experimentally obtained internal friction presented above can be compared with the values calculated using the Eq.1 and Eq. 2. We have directly measured the sample thickness, (0.305± 0.005) and (0.183±0.005) mm respectively, and calculated the material density (both taken as constant in function of temperature). The measured temperature dependent resonant frequency $f(T)$ and calculated Young's modulus $E(T)$ are also available from our experiments. The temperature dependent values of thermal expansion coefficient $\alpha(T)$, specific heat capacity $c(T)$ and thermal conductivity $\lambda(T)$ were taken from literature [4, 5, 6].

Fig. 2 shows the values used for calculation superposed to the original literature graphs. The compression and tension during the sample vibration are occurring along the length of the sample and the samples measured had the c direction of hexagonal crystal structure oriented along the sample thickness. Therefore, the thermal expansion coefficient is taken along a-axis from reference [4].

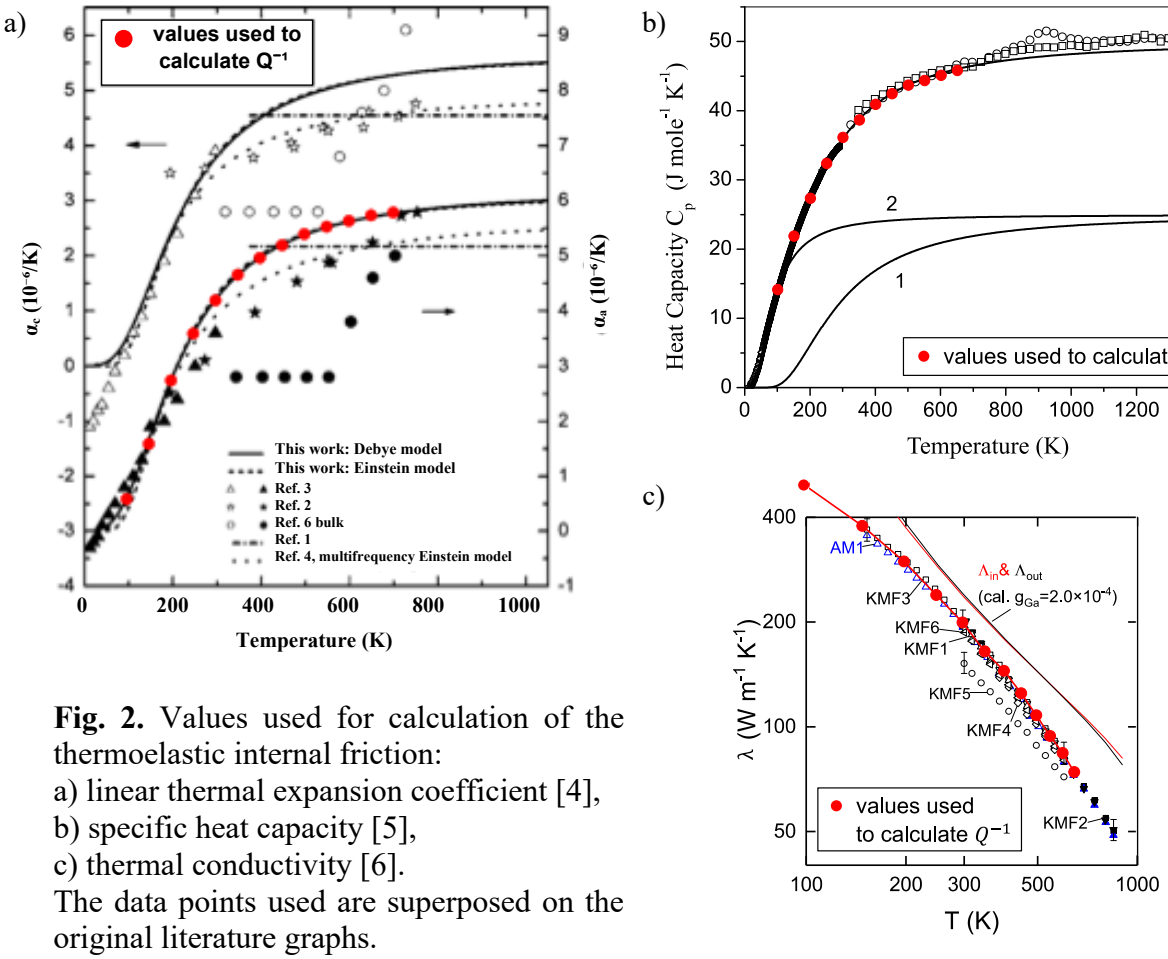
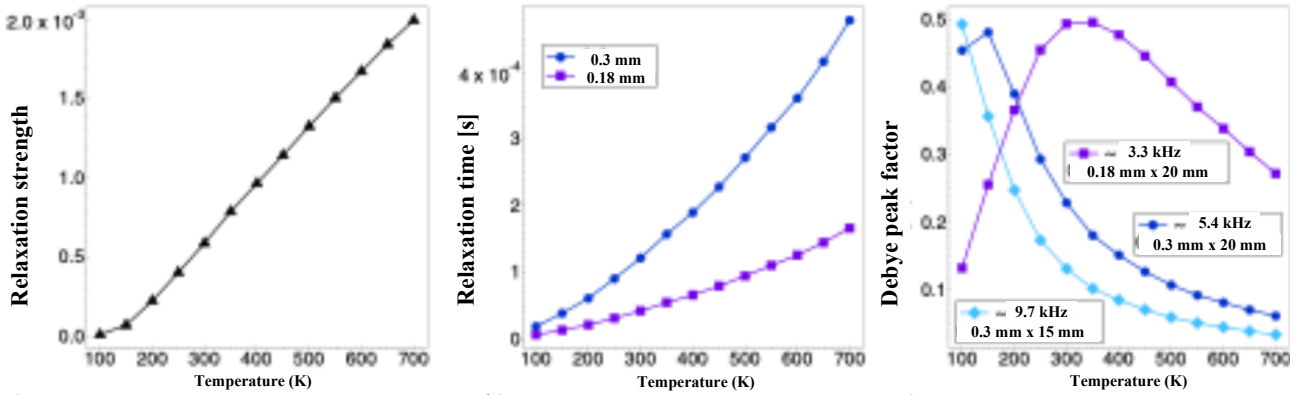


Fig. 2. Values used for calculation of the thermoelastic internal friction:
a) linear thermal expansion coefficient [4],
b) specific heat capacity [5],
c) thermal conductivity [6].
The data points used are superposed on the original literature graphs.

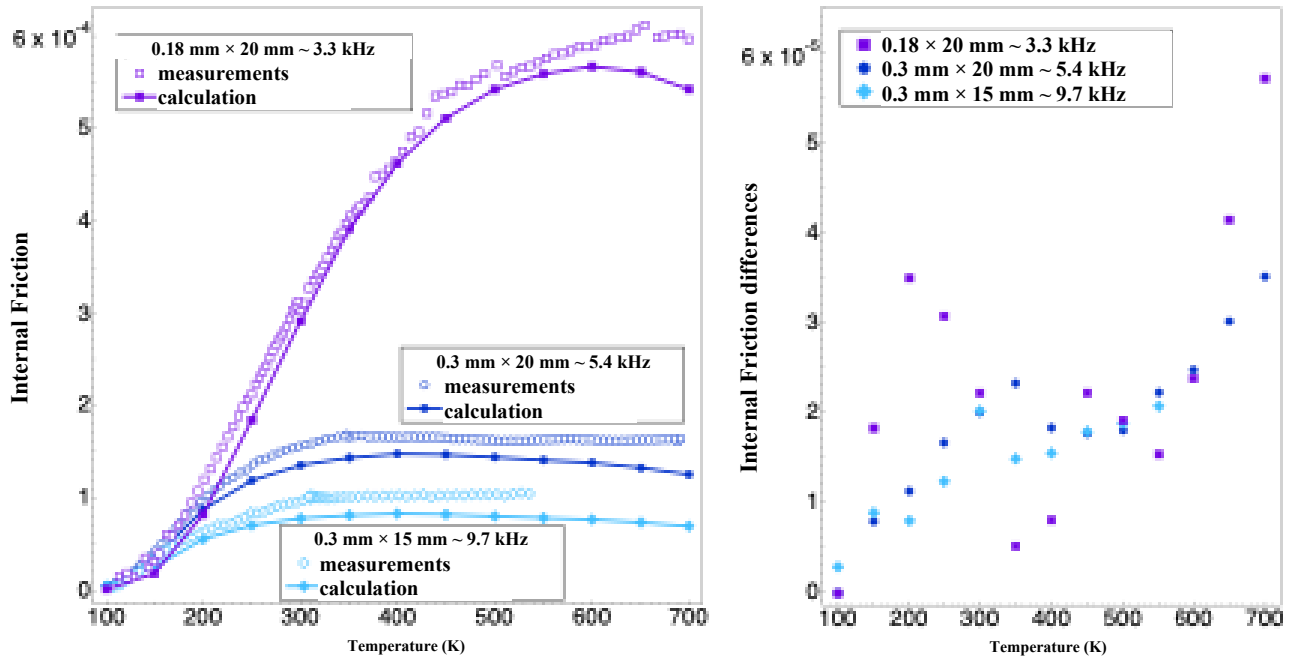
Calculated individual components to the thermoelastic damping are presented in Fig. 3. The relaxation strength Δ in Eq. 1 is plotted in Fig. 3a. It does not depend on the sample dimensions nor the resonant frequency and is therefore the same for all samples. The relaxation time calculated using Eq. 2 depends on the sample thickness and is shown in Fig. 3b. Finally, the Debye peak factor, given in the brackets of Eq. 1, is presented in Fig. 3c. It depends both on the sample thickness through the relaxation time, and the frequency which changes also with the sample length and is therefore different for each sample.



a) b) c)

Fig. 3. Calculated components of the thermoelastic damping as a function of temperature. a) Relaxation strength. b) Relaxation time. c) Debye peak factor.

The total calculated thermoelastic temperature dependent internal friction is presented together with the measured one in Fig. 4a. There are no adjustable (fitted) parameters used for the calculations. The agreement between the theory and the experiment is globally very good. The difference between the measured and the calculated values is presented in Fig. 4b.



a) b)

Fig. 4. a) Comparison between the measured internal friction for three GaN samples with different dimensions and resonant frequency with the calculated thermoelastic damping. b) Difference between the measured and the calculated internal friction for each sample.

Discussion

The room temperature value of Young's modulus is found to be (287±10) GPa. The uncertainty is mostly due to the precision of the dimensions, particularly of the sample thickness, as the samples were not perfectly flat. The obtained Young's modulus value corresponds to the one found for the thin films by Tsai et al. [7]. The Young's modulus monotonously decreases upon heating. The sample density and dimensions are often taken to be temperature independent for modulus calculation using Eq. 3. The effect of thermal expansion on the Young's modulus was verified using the temperature dependent coefficient of the thermal expansion [4]. The biggest difference is observed at the highest temperature, but as the modulus was found to be less than 0.2% lower in this extreme case, the correction is not applied in our calculations.

The measured internal friction depends very strongly on the sample dimensions and therefore the resonant frequency (Fig. 1b), but not in a way characteristic for defect-related thermally activated mechanisms. For those, the relaxation time follows the Arrhenius law and decreases with temperature. Consequently, the characteristic relaxation maxima shift to higher temperatures when the measurement frequency increases. Fig. 1b shows the opposite behaviour that can be attributed to the thermoelastic damping. The relaxation time calculated for this mechanism (Fig. 3b) increases with temperature. The increase is caused both by the increase of the specific heat capacity c (Fig. 2b) and the decrease of the thermal conductivity λ (Fig. 2c) for higher temperatures. The Debye peak factor (Fig. 3c) exhibits a clear maximum for the lowest frequency of ~ 3.3 kHz. The maximum shifts to lower temperatures for higher frequencies. As the relaxation strength Δ also increases with temperature (Fig. 3a), the thermoelastic damping exhibits a very broad maximum as a function of temperature, looking like a high temperature plateau.

The thermoelastic damping contribution calculated as a function of temperature using the literature data and measured quantities is in a very good agreement with the general shape of the experimental curves in almost the whole temperature range (Fig. 4a) and it is clearly the dominant dissipation mechanism. The difference between the measured and the calculated values (Fig. 4b) is of the order of 10^{-5} and it starts to increase more significantly only above 600 K. For temperatures below 600 K, this difference does not seem to be dependent on frequency, there is only some more dispersion at low temperatures for the measurement for the thinner sample at 3.3 kHz. It can be concluded that the other potential thermally activated energy dissipation mechanisms, like the point defect relaxation for example, could begin to contribute to internal friction only at temperatures higher than 600 K for the frequencies in the kHz range.

For lower temperatures, the other potential damping mechanisms that could explain the difference between the measured and the thermoelastic damping, also observed in the case of silicone cantilevers [8], include the damping from residual gas in the chamber and surface loss due to the surface roughness. There is also a potential contribution to the damping from the suspension wires. They were supposed to be placed exactly in the nodal position of the first flexural vibration mode, but due to their non-infinitesimal thickness (15 μm), the uncertainty of the sample length and manual centering when mounting the sample, a certain low contribution to the damping cannot be excluded.

The small differences found between the measured values of Q^{-1} and the model raise the question of the influence of the precision of the parameters taken from literature that are then used to calculate the thermoelastic damping. Namely, if the value of the thermal expansion coefficient α and/or the specific heat capacity c change of only a few percents, this would suffice to further reduce the discrepancy between the model and the measurements. The dispersion of the published experimental values (Fig. 2a and 2b) is not far from this estimate.

Conclusion

The temperature dependence of Young's modulus and internal friction in bulk GaN single crystal were measured for the first time. Young's modulus is found to monotonously decrease upon heating from (287±10) GPa at room temperature to a value at 700 K 2% lower. The internal friction is strongly dependent on the sample dimensions and the oscillation frequency. It is compared to the thermoelastic

damping calculated using the experimentally obtained values for sample thickness, density, frequency and Young's modulus, as well as the temperature dependent parameters found in literature: linear thermal expansion coefficient, specific heat capacity and thermal conductivity. The agreement is very good for temperatures below 600 K for all frequencies. The obtained experimental results are therefore a very convincing confirmation of the validity of the Zener model in a large temperature range. The difference between the thermoelastic damping and the measured internal friction is relatively low and independent of frequency. The thermoelastic effect is clearly the dominant energy dissipation mechanism since the crystal defects in GaN such as point defects or dislocations are not mobile for temperatures lower than 600 K and frequencies in the kHz range.

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