Effect of dislocations on thermal conductivity of GaN layers

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We report calculation of the lattice thermal conductivity in wurtzite GaN. The proposed model is material specific and explicitly includes phonon relaxation on threading dislocations and impurities typical for GaN. We have found that a decrease of the dislocation density by two orders of magnitude in GaN leads to a corresponding increase of the thermal conductivity from 1.31 to 1.97 W/cm K. This theoretical prediction is in very good agreement with experimental data obtained from scanning thermal microscopy. The developed model can be used for thermal budget calculations in high-power density GaN devices. © *2001 American Institute of Physics.*
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Proposed applications of GaN-based devices as laser diodes, microwave power sources, and ultrahigh power switches rely heavily on the possibility of removing high density of excess heat from the device active area. The latter, in turn, depends on the thermal conductivity values in GaN and related compounds. The first measurements of the thermal conductivity (K) of GaN films grown by hydride vapor phase epitaxy revealed a rather low value of about 1.3 W/cm K at room temperature.[1] For comparison, the thermal conductivity of Si is 1.45 W/cm K at $T=300$ K. Recently, Asnin *et al.*[2] have performed high spatial resolution measurements on lateral epitaxial overgrowth (LEO) samples grown on sapphire and found values of 1.7–1.8 W/cm K. Using the same scanning thermal microscopy (SThM) technique, Florescu *et al.*[3,4] determined that the thermal conductivity of the overgrown LEO regions is nearly two times higher (2.1 W/cm K) than that between stripes, e.g., in the vicinity of the SiN_x mask. Based on the qualitative considerations Florescu *et al.*[4] suggested that the variation of thermal conductivity in LEO GaN/sapphire (0001) samples is due to the change in the threading dislocation line density.

In this letter we report a theoretical calculation of the lattice thermal conductivity in wurtzite GaN. Our first goal is to develop a model for calculating the thermal conductivity in GaN films characterized by realistic material parameters specific for a given growth technique. The second goal is to qualitatively investigate a hypothesis that a change in the dislocation line density can be responsible for the observed large variation in thermal conductivity. Despite the significant practical importance of knowledge of thermal transport in GaN materials and a number of recent experimental reports on thermal conductivity values, the theoretical investigation of the subject lagged behind. To date, the available theoretical models of thermal conductivity in GaN are limited to calculations of the *intrinsic* theoretical limit[5,6] that do not take into account defects and impurities.

Heat in semiconductors is mostly carried by *acoustic* phonons. Thus, here we neglect the electronic component of thermal conductivity. For isotropic materials, the expression for the lattice thermal conductivity can be written as[7,8]

$$K=\frac{1}{3}\sum_{\mathbf{q},j} C_j(\mathbf{q})V_j^2(\mathbf{q})\tau_{C,j}(\mathbf{q}) \approx \frac{k_B}{2\pi^2 V}\left(\frac{k_B}{\hbar}\right)^3 T^3\int_0^{\theta_D/T}\frac{\tau_C x^4 e^x}{(e^x-1)^2}dx. \quad (1)$$

Here k_B is the Boltzmann constant, $\hbar$ is the Planck constant, T is the absolute temperature, θ_D is Debye temperature, subscript j denotes a specific phonon polarization type, τ_C is the combined phonon relaxation time, $\mathbf{q}$ is the phonon wave vector, ω is the phonon frequency, C is the specific heat per unit volume, and $x=\hbar\omega/k_BT$. Equation (1) is derived using Debye's phonon density-of-states and under the assumption of linear phonon dispersion $\omega=V_jq$ with spherical $\omega=$constant surface in $\mathbf{q}$ space. It is accurate for $T\geqslant 300$ K and the case when resistive processes are dominant. The latter assumption is valid for GaN with characteristic high defect densities. The sound velocity V_j is averaged over all phonon angles, then the mean velocity V is calculated for all phonon polarization types from the expression $3/V^3=1/V_L^3+2/V_T^3$. Detailed analysis of sound velocity in GaN for longitudinal (L) and transverse (T) phonons has been given by Deguchi *et al.*[9] Here, we use the direction-independent sound velocity because heat in the reference SThM experiments propagates in all directions (see inset of Fig. 1). Velocities V_L and V_T are related to the elastic constants $C_{L,T}$ through $V_{L,T}=\sqrt{C_{L,T}/\rho}$ and $\mu=V_T^2\cdot\rho=C_T$ where ρ is the mass density and μ is the shear modulus.

In order to obtain the intrinsic thermal conductivity limit, we need to assume that thermal resistance is only due to crystal anharmonicity and calculate the phonon relaxation rate $1/\tau_U$ in three-phonon Umklapp scattering processes. At high temperature this rate is given as[7]

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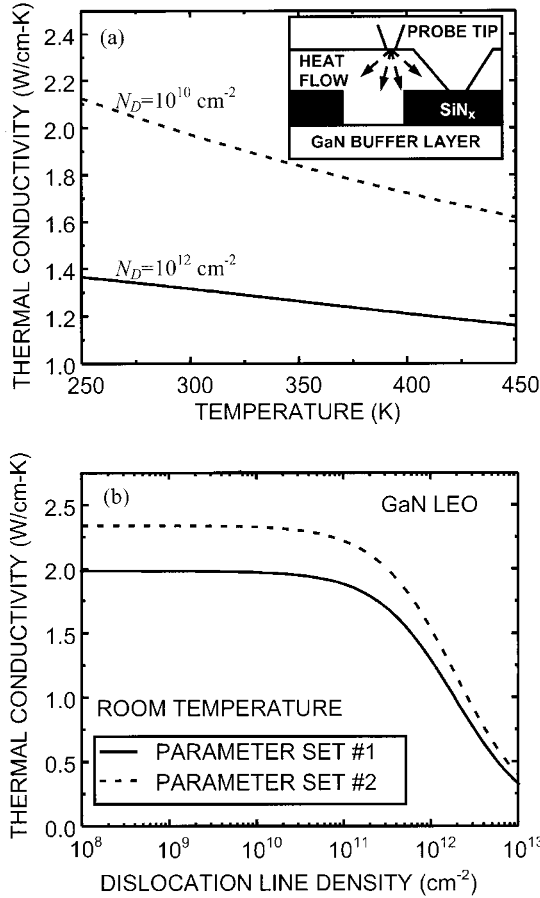

FIG. 1. Thermal conductivity of GaN as a function of temperature (a), and the dislocation line density (b). Inset (a) shows structure of the LEO grown GaN sample and direction of the heat flow. The results in (b) are obtained for two different sets of material parameters summarized in Table I.

$$\frac{1}{\tau_U}=2\gamma^2\frac{k_BT}{\mu V_0}\frac{\omega^2}{\omega_D}, \tag{2}$$

where γ is the Gruneisen anharmonicity parameter, and V_0 is the volume per atom. Since there are four atoms in a unit cell, $V_0=|\mathbf{a_1}\cdot(\mathbf{a_2}\times\mathbf{a_3})|/4=a^2c3^{1/2}/8$. Here a and c are lattice constants, and $\mathbf{a_1}$, $\mathbf{a_2}$, and $\mathbf{a_3}$ are primitive vectors for wurtzite GaN. Inserting Eq. (2) into Eq. (1) with $\tau_C=\tau_U$ we can now numerically evaluate Umklapp-limited thermal conductivity of GaN.

There is significant discrepancy in values of material parameters reported for GaN. Due to this reason we evaluate K for two distinctively different sets of material parameters reported in Refs. 10 and 11, which are summarized in Table I. As one can see from the table, the values of the intrinsic thermal conductivity obtained from our model are consistent with previously reported theoretical limit of GaN thermal conductivity of $K=4.1$ W/cm K.[5] The experimentally measured values of K in GaN are much smaller than the calculated theoretical limit because imperfections and impurities in GaN lattice scatter acoustic phonons thus introducing additional thermal resistance. From the second-order perturbation theory the phonon relaxation rate on point defects can be written as[7]

TABLE I. Material parameters and simulation results.

Material parameters		Set I	Set II
Lattice constant	a (Å)	3.189[a]	3.189[a]
Lattice constant	c (Å)	5.185[a]	5.185[a]
Gruneisen parameter	γ	0.74[b]	0.74[b]
Density	ρ (kg/m^3)	6150	6150
Longitudinal elastic constant	C_L (GPa)	265	293
Transverse elastic constant	C_T (GPa)	44.2	81
Debye temperature	θ_D (K)	1058	830[b]
Hydrogen impurity	H (l/cm^3)	4×10^{18}	4×10^{18}
Carbon impurity	C (l/cm^3)	1.5×10^{18}	1.5×10^{18}
Oxygen impurity	O (l/cm^3)	2×10^{18}	2×10^{18}
Silicon impurity	Si (l/cm^3)	8×10^{17}	8×10^{17}
Intrinsic thermal conductivity	K (W/cm K)	3.70	3.44

[a]Estimated from Ref. 13.
[b]Estimated from Ref. 5.

$$\frac{1}{\tau_M}=\frac{V_0\omega^4}{4\pi V^3}\Gamma=\frac{V_0\omega^4}{4\pi V^3}\sum_i f_i\left(1-\frac{M_i}{\bar{M}}\right)^2, \tag{3}$$

where f_i is the relative concentration of the ith atoms, $\bar{M}=\Sigma_i f_i M_i$ is the average atomic mass, M_i is the mass of the ith impurity atom or defect, and Γ is the measure of the strength of the point-defects scattering. In Eq. (3) we neglect contributions from vacancies, the host atom–impurity atom linkage anharmonicity and from the impurity atom volume difference. These contributions are usually small and in the simulation procedure can be compensated by some increase in the effective concentration of mass difference term.

Characteristic residual impurities in GaN grown by metalorganic chemical vapor deposition (MOCVD) or molecular beam epitaxy (MBE) are H, O, C, and Si.[12,13] H is a component of many gases used in GaN growth, such as SiH_4, H_2, NH_3. It was observed that atomic H diffuses rapidly in GaN material, especially when dopants are present. The source of O in GaN is usually NH_3 precursor used in MOCVD growth, the residual water vapor in MBE chambers or O impurities leached from the quartz containment vessel often employed in N_2 plasma sources.[12] The source of C impurity in GaN can be the metalorganic precursor $(CH_3)_3Ga$.[14] Finally, the source of Si impurity in LEO grown samples can be diffusion from SiN_x mask or n-type doping with SiH_4 or Si_2H_6. It is known that even the samples without intentional n-type doping have Si concentration comparable to that of C and H. Concentrations of impurities used in simulation are shown in Table I.

Phonon scattering on dislocations is described by a combined term that includes scattering from the cylindrical imperfection, e.g., core dislocation, and from the long-range strain field, which includes screw and edge dislocation components $\tau_D^{-1}=\tau_{\rm core}^{-1}+\tau_{\rm screw}^{-1}+\tau_{\rm edge}^{-1}$. In the case when dislocation lines are perpendicular to the temperature gradient the scattering rates for the dislocation core and long-range strain field are given by[15]

$$\frac{1}{\tau_{\rm core}}=N_D\frac{V_0^{4/3}}{V^2}\omega^3 \quad \text{and} \quad \frac{1}{\tau_{\rm screw}}=0.06N_Db^2\gamma^2\omega, \tag{4}$$

where N_D is the dislocation line density, and b is the magnitude of Burgers vector. The edge dislocation $\tau_{\rm edge}^{-1}$ is given by

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the same expression as $\tau_{\mathrm{screw}}^{-1}$ but with different values of the Burgers vector. Dislocation lines in LEO GaN samples are predominantly vertically arranged although there is no strict order and some bending and disorder are always present.[16] The dislocations in GaN regrown on grooves tend to propagate off the c axis so that an area with significantly reduced dislocation density is formed above the grooves. Since the heat in SThM measurement technique propagates in all directions (see inset of Fig. 1), we multiply rates in Eq. (4) by 0.55 to account for randomness in the mutual direction of heat propagation and dislocation line.[7,15]

The combined relaxation rate $1/\tau_C$ in Eq. (1) is then calculated as a function of ω using the expression $1/\tau_C = 1/\tau_U + 1/\tau_M + 1/\tau_D$. We calculate the lattice thermal conductivity for two material parameter sets in Table I. Figures 1(a) and 1(b) show K in GaN as a function of temperature and dislocation line density, respectively. The inset illustrates the structure of the LEO GaN sample and the direction of heat flow. As one can see, at $T=300$ K, a two order of magnitude drop in the dislocation line density (from 10^{12} to 10^{10} cm^{-2}) brings about considerable increase in the lattice thermal conductivity from 1.31 to 1.97 W/cm K. The latter agrees well with recent experimental observations[2–4] and explains earlier measured low values of thermal conductivity.[1] Most recent SThM measurements demonstrate a variation of K from 1.9 to 1.1 W/cm K in LEO GaN/sapphire samples,[17] which is consistent with our calculations. The conclusion that can be made from Fig. 1(b) is that for N_D on the order of $10^{10}-10^{11}$ cm^{-2}, which is typical for regular grown GaN,[13,14,16–18] dislocations play an important role in thermal resistance $1/K$ at room temperature. As dislocation density is reduced below 10^8 cm^{-2} by some special means,[16] their presence does not affect the thermal conductivity, which is in this case mostly defined by intrinsic crystal properties and point impurities. In conclusion, we proved that experimentally observed variation of thermal conductivity in LEO GaN/sapphire samples could be attributed to dislocation inhibition in overgrown regions. The developed model for thermal conductivity in wurtzite GaN can be used for thermal budget calculation in the high-power density GaN devices.

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[1] E. K. Sichel and J. I. Pankove, J. Phys. Chem. Solids **38**, 330 (1997).
[2] V. M. Asnin, F. H. Pollak, J. Ramer, M. Schurman, and I. Ferguson, Appl. Phys. Lett. **75**, 1240 (1999).
[3] 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**, 1464 (2000).
[4] D. I. Florescu, V. M. Asnin, and F. H. Pollak, Compound Semicond. **7**, 62 (2001).
[5] A. Witek, Diamond Relat. Mater. **7**, 962 (1998).
[6] G. A. Slack, J. Phys. Chem. Solids **34**, 321 (1973).
[7] P. G. Klemens, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1958), Vol. 7.
[8] R. Berman, in *Thermal Conduction in Solids* (Clarendon, Oxford, 1976), p. 37.
[9] T. Deguchi, D. Ichiryu, K. Toshikawa, K. Sekiguchi, T. Sota, R. Matsuo, T. Azuhata, M. Yamaguchi, T. Yagi, S. Chichibu, and S. Nakamura, J. Appl. Phys. **86**, 1860 (1999).
[10] V. W. L. Chin, T. L. Tansley, and T. Osotchan, J. Appl. Phys. **75**, 7365 (1994).
[11] R. B. Schwarz, K. Khachataryan, and E. R. Weber, Appl. Phys. Lett. **70**, 1122 (1997).
[12] S. J. Pearton, J. C. Zolper, R. J. Shul, and F. Ren, J. Appl. Phys. **86**, 1 (1999).
[13] S. Nakamura, S. Pearton, and G. Fasol, in *The Blue Laser Diode: The Complete Story* (Springer, Berlin, 2000).
[14] R. Zhang and T. F. Kuech, Appl. Phys. Lett. **72**, 1611 (1998).
[15] P. G. Klemens, Proc. Phys. Soc., LXVIII **12-A**, 1113 (1955).
[16] M. Ishida, M. Ogawa, K. Orita, O. Imafuji, M. Yuri, T. Sugino, and K. Itoh, J. Cryst. Growth **221**, 345 (2000).
[17] F. H. Pollak (unpublished).
[18] S. D. Lester, F. A. Ponce, M. G. Craford, and D. A. Steigerwald, Appl. Phys. Lett. **66**, 1249 (1995).

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