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High spatial resolution thermal conductivity of bulk ZnO (0001)

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We measured high spatial/depth resolution 300 K thermal conductivity κ of the Zn and O surfaces of two bulk *n*-type ZnO (0001) samples, grown by a vapor-phase transport method, using scanning thermal microscopy (SThM). The thermal investigation was performed in both point-by-point (~ 2 μm resolution) and area-scan modes. On the first sample $\kappa=1.16\pm 0.08$ (Zn face)/ 1.10 ± 0.09 (O face) W/cm K while for the second material $\kappa=1.02\pm 0.07$ (Zn face)/ 0.98 ± 0.08 (O face) W/cm K. These are the highest κ values reported on ZnO. A correlation between SThM area-scan readings and surface topography was established by simultaneously performing atomic force microscopy scans. The influence of surface roughness on the effective thermal conductivity (i.e., heat flow) is discussed. © 2002 American Institute of Physics. [DOI: 10.1063/1.1426234]

ZnO is a wide band gap (3.5 eV) II–VI semiconductor, which has been used in many applications such as piezoelectric transducers,¹ varistors,¹ and laser diodes.² Recently, ZnO has been recommended as an attractive alternative to both sapphire and SiC as substrates for GaN growth. When growing GaN on (0001) sapphire substrates, one has to cope with a 16% lattice mismatch.³ SiC is better suited as a substrate material, but is costly and also generates stacking mismatch boundaries in device structures, which shorten the lifetime of the device. For ZnO the lattice mismatch is only 2.2% for GaN and zero for $\text{In}_{0.22}\text{Ga}_{0.78}\text{N}$.³ Since most of these applications are related to high power electronic and optoelectronic devices it is essential to understand the thermal characteristics of ZnO. The thermal conductivity κ of solid materials consists of both lattice and electronic contributions. The former is a function of the mean free path of the phonons and hence is determined by both intrinsic (anharmonic phonon–phonon scattering) and extrinsic (phonon–defect) scattering. For carrier concentration $\leq 10^{19}\text{ cm}^{-3}$ the electronic thermal conductivity is negligible in relation to the lattice portion. The quantity κ is of importance from both fundamental (basic mechanism of phonon propagation) and applied (device modeling, sample quality) perspectives.

We report high spatial/depth resolution κ measurements at 300 K of the Zn and O faces of two *n*-type ZnO (0001) samples fabricated by a vapor-phase technique. The thermal investigation was performed in two ways: point-by-point (~ 2 μm resolution) and area scans. The area scans are instrumentation limited to approximately $120\text{ }\mu\text{m}\times 120\text{ }\mu\text{m}$ for one scan, while the point-by-point measurements can be extended to full 8 in. wafers. The point-by-point measurements on sample A reveal $\kappa=1.16\pm 0.08$ (Zn face)/ 1.10 ± 0.09 (O face) W/cm K, while for sample B $\kappa=1.02\pm 0.07$ (Zn face)/ 0.98 ± 0.08 (O face) W/cm K. These are approximately twice higher than the previously reported value of ~ 0.48 W/cm K.⁴ A correlation between effective κ values from the scanning thermal microscopy (SThM) scans and topography was established by simultaneously performing atomic force microscopy (AFM) scans.

The samples used in this study were two 6 mm $\times$ 6 mm *n*-type ZnO (0001) pieces cut from full 2 in. wafers grown by a vapor-phase technique described elsewhere.¹ The Zn and O faces were produced by a proper chemomechanical polishing method.¹ The carrier concentration and mobility were previously measured by the temperature-dependent Hall-effect technique.¹ The sample parameters are shown in

TABLE I. Summary of the sample parameters.

Sample	Thickness (μm)	Mobility ($\text{cm}^2\text{ V}^{-1}\text{ s}^{-1}$)	Carrier		
			concentration (cm^{-3})	κ -Zn side (W/cm K)	κ -O side (W/cm K)
A	650	211	5.2×10^{16}	1.16 ± 0.08	1.10 ± 0.09
B	565	227	6.6×10^{16}	1.02 ± 0.07	0.98 ± 0.08

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FIG. 1. (a) AFM and (b) SThM scans on a $16\ \mu\text{m} \times 16\ \mu\text{m}$ area of the Zn side of sample B. Note the correlation between the SThM and AFM scan features.

Table I. The high spatial resolution thermal conductivity measurements were made using a ThermoMicroscopes Auto-Probe M5 SThM/AFM.⁵ A calibration procedure has been developed making it possible to determine absolute values of κ . The output voltage V_{out}^2 has been measured for a series of samples of known thermal conductivity.^{6,7} The resultant linear relation between V_{out}^2 and κ provides the calibration. AFM scans were acquired concomitantly with SThM area scans using the same tip element.

Table I shows both the Zn and O side thermal conductivity results using the point-by-point type measurement technique on both samples. Approximately 15–20 points randomly distributed over the $6\ \text{mm} \times 6\ \text{mm}$ surface area were investigated. The variation is the standard deviation.

AFM and SThM scans are presented in Figs. 1(a) and 1(b), respectively, on a $16\ \mu\text{m} \times 16\ \mu\text{m}$ area on the Zn side of sample B. Similar scans were acquired on the O side, and also on both sides of sample A, and will not be presented herein. SThM scans provide information only on the relative variations of the thermal conductivity.⁸ Note the correlation between the SThM and AFM scan features. SPMLAB V5.01⁵ was used on the SThM scan to display ΔV_{out}^2 (variations in V_{out}^2), which are proportional to the changes in the effective thermal conductivity. Based on our calibration, the ΔV_{out}^2 range observed in Fig. 1(b) corresponds to variations of about 6%–7% in κ . This is consistent with the deviations reported in Table I for the point-by-point type measurements.

The following question arises: are these small variations in the observed V_{out}^2 an intrinsic material property, i.e., changes in the actual thermal conductivity, or just the effect of surface topography? Several factors, such as the contact resistance between tip and probe, the temperature distribution along the tip, the contact force, the surrounding temperature, humidity, and gas are influencing the voltage reading when the tip is in contact with the specimen.^{8,9} Since the calibration and measurements are performed under the exact same conditions, the only factor that differs from sample to sample is the surface topography. For a perfectly flat surface

the contact between the probe tip (radius of curvature $\sim 1\ \mu\text{m}$)^{6,7} and the surface is very small, with spatial resolutions higher than the actual tip dimension being reported for SThM scans.¹⁰ For rough surfaces, while scanning or positioning, the tip could impinge on a valley-like feature [Fig. 2(a)], or a hillock [Fig. 2(b)]. We performed a calculation of the effective heat flow for topographical features with radius of curvature r_c ($\pm$ for hillock/valley), and height/depth h ($|h| \leq |r_c|$), assuming in-plane symmetry (see Fig. 2). For a perfectly flat surface $r_c \rightarrow \infty$, $h \rightarrow 0$. As seen in Fig. 1(a) the height variations when scanning across the sample are in the 0–100 nm range. For $20\ \text{nm} < |h| < 100\ \text{nm}$ and $|r_c| \leq 10\ \mu\text{m}$ the results show that the difference in heat flow in relation to the flat surface is about 7%–12%, being negative/positive for hillock/valley. For $|r_c| > 10\ \mu\text{m}$ this difference is even less. Hence, a valley will lead to an increased thermal signal, and vice versa. Thus topography can have an inverted effect on the thermal conductivity measurements, as shown in Fig. 1.

FIG. 2. Geometry of SThM/AFM tip and sample surface interaction. The tip can be (a) in a valley, or (b) on top of a hillock. The parameter r_c is the hillock/valley radius of curvature, and h is the height/depth from the sample surface.

The noticeable difference in spatial resolutions between the AFM and SThM scans is related to changes in the effective heat flow between the scanning probe and sample surface, which are more significant on the voltage, i.e., thermal readings, than on AFM topography readings. Features as small as $0.5\ \mu\text{m}$ and up to $\sim 3\ \mu\text{m}$ wide are clearly resolved by both the AFM and SThM scans, as shown in Fig. 1. However, the experimental voltage variations ΔV_{out}^2 correspond to only 6%–7% changes in the thermal conductivity. In this case κ variations are probably the effect of topography. Higher variations ($>12\%$) in the SThM V_{out}^2 reading (proportional to κ) for AFM topographical features with heights $<100\ \text{nm}$ would be an indication of intrinsic thermal conductivity variations across the area under investigation.

In conclusion, by using SThM we have performed high spatial/depth resolution thermal conductivity measurements at 300 K of two *n*-type ZnO (0001) samples grown by a vapor-phase transport method. Point-by-point and area-scan type measurements were employed. On the Zn(O) side we found $\kappa = 1.16 \pm 0.08$ (1.10 ± 0.09) W/cm K on sample A, and $\kappa = 1.02 \pm 0.07$ (0.98 ± 0.08) W/cm K on sample B. To date these are the highest reported thermal conductivity values on ZnO. A correlation between the surface topography and thermal conductivity variations was established by acquiring the AFM and SThM scans concomitantly during the area scan.

The effects of surface roughness on effective κ values, i.e., heat flow, were discussed in terms of tip–surface geometry.

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