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Submicron Mapping of Thermal Conductivity of Thermoelectric Thin Films

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A tool for evaluating thin-film thermal conductivity to submicron spatial resolution has been developed. The micro-instrumentation utilizes the thermoreflectance (TR) technique to characterize thermal conductivity and material uniformity. The instrument consists of a heating element for creating temperature gradients and an Invar bar with *in situ* temperature monitoring for heat flux measurements. The thin-film sample is sandwiched between the heater and Invar bar while a microscope is used to direct light onto a cross-section of the sample and reflected light is collected with a camera. By using this technique, we can achieve submicron spatial resolution for thermal conductivity and eliminate contributions from thermal contact resistance, thereby also eliminating the need for sample preparation other than cleaving. The method offers temperature resolution of 10 mK, spatial resolution of 200 nm, and thermal conductivity measurement with 0.01 ± 0.001 W/mK resolution. The thermal conductivity of a 0.6% ErAs:InGaAlAs thermoelectric (TE) element, prepared by molecular beam epitaxy (MBE) growth, obtained with the new instrument is 2.3 W/mK, while the average thermal conductivity obtained with the 3-omega method is 2.5 W/mK. Energy-dispersive x-ray (EDX) spectroscopy is also used to prove that the elemental composition has uniformity consistent with the material variation observed by the TR technique. Moreover, a temperature profile across a 0.6% ErAs:InGaAlAs TE element on InP substrate is imaged. Two different slopes, corresponding to different thermal conductivities, have been observed, showing that the thermal conductivity of the TE element is lower than that of the InP substrate as expected.

Key words: Thermoelectric, thermal conductivity, thermoreflectance

INTRODUCTION

There are various approaches to measuring thin-film thermal conductivity, such as the 3-omega method,[1–3] scanning thermal microscopy (SThM),[4–7] and photothermal reflectance techniques.[8,9] The 3-omega method requires a metal heater that also acts as a thermometer, being fabricated on top of a sample. For electrically conductive samples, additional dielectric materials are also required before the heater/thermometer fabrication process. This method requires sample preparation and measures an average value of the material property. In contrast to the 3-omega method, SThM is a nanoscale thermometer that maps the local temperature and thermal conductivity. Two dissimilar inner and outer conductors separated by an insulator except at the tip (~100 nm) form the SThM thermocouple junction. The relatively slow rate of scanning during thermal imaging often leads to drift in the image. Moreover, the heat conduction between the tip and surface defines the spatial resolution. The thermal design of the probe is critical since it drives the measurement uncertainty. Apart from its fast response, the photothermal reflectance technique is

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a noncontact optical method to evaluate temperature variations. It uses a heating laser to create periodic sample heating. The change in reflectivity due to the periodic heating by the pump beam is detected using a probe laser and measured by lock-in detection. The photothermal reflectance technique is capable of directly measuring thermal decay in the surface, but it requires knowledge of the specific heat of the sample. In this work we have developed a tool for evaluating thin-film thermal conductivity to submicron spatial resolution. The method is noncontact and offers temperature resolution of 10 mK, spatial resolution of 200 nm, and thermal conductivity measurement with 0.01 ± 0.001 W/mK resolution.[10–12] This micro-instrumentation utilizes the TR technique to characterize thermal conductivity and material uniformity. Instead of applying current to the sample (Joule heating or Peltier cooling),[11] temperature modulations are created by an external heating element. It operates at relatively low frequency compared with the time-domain TR technique.[13] There is no sample preparation needed other than cleaving, and this method eliminates contributions from thermal contact resistances. It gives a map of the thermal conductivity distribution. Compared with the photothermal reflectance technique,[8,9] it can image over a large area simultaneously, in addition to the simplicity of the optical setup. However, it requires calibration of the TR coefficients of the sample.

EXPERIMENTAL PROCEDURES

We have developed micro-instrumentation utilizing the TR technique to characterize thermal conductivity and material uniformity. The instrument, shown in Fig. 1, consists of a heating element for creating temperature gradients and an Invar bar with *in situ* temperature monitoring for heat flux measurements. Invar was chosen due to its low thermal expansion coefficient and thermal conductivity. The dimensions of the Invar bar are set to have thermal resistance on the same order of magnitude as that of the sample of interest. With knowledge of the dimensions, thermal conductivity, and temperature gradient obtained by thermocouple, the heat flux across the Invar bar, and therefore the sample, can be calculated. The thin-film sample is sandwiched between the heater and Invar bar (measurement bar) while a microscope is used to direct light onto a cross-section of the sample and reflected light is collected with a camera. The changes in reflectivity (ΔR) caused by changes in surface temperature (ΔT), as described by the relationship $\Delta T = \Delta R/\kappa R$, where κ is the material- and wavelength-dependent TR coefficient and R is the reflectivity at room temperature. The TR coefficient is typically small (10^{-3} K^{-1} to 10^{-5} K^{-1}); therefore, a lock-in technique is used to improve signal sensitivity. A steady source of fluorescence illumination (wavelength: 450 nm to 500 nm) is used to illuminate the sample, and the

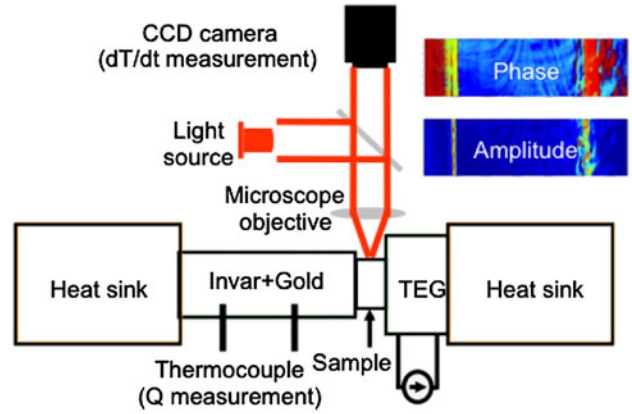

Fig. 1. Schematic diagram of micro-instrumentation for thermal conductivity measurements.

image is projected onto a 650 × 400 pixel charge-coupled device (CCD) array microscope. The temperature modulation of the sample is created by modulating the TE heating element using a sinusoidal current at frequency 1ω. The CCD camera then takes pictures of the sample through an optical bandpass filter (wavelength: 460 nm to 480 nm). To detect the temperature heating occurring at frequency of 1ω, the camera takes images of the device at trigger frequency of 4ω, phase-locked to the thermal excitation. The spatial resolution is limited by the microscope and microscope objective according to the diffraction limit as well as any long-term drift that is present in the image train. The noise in the temperature profiles is not set by the instrument, but rather by scratches and defects on the surface of the sample. A smooth cleaved surface is crucial for TR measurements. By using this technique, we can achieve high spatial resolution for thermal conductivity and eliminate contributions from thermal contact resistance, but also eliminate the need for sample preparation other than cleaving.

RESULTS AND DISCUSSION

The instrument was utilized to examine (1) the thermal conductivity of a 60-μm ErAs:InGaAlAs TE element and (2) the temperature profile across a 5-μm ErAs:InGaAlAs TE element on a 45-μm InP substrate. ErAs:InGaAlAs was chosen due to its tendency to cleave along definite smooth planar surfaces determined by its crystal structure, which is crucial for TR measurements. The 60-μm sample has uniform material composition and was used to validate the accuracy of the measurement. The thermal conductivity obtained by the 3-omega method was 2.5 W/mK.[13] This thermal conductivity range is representative for TE elements and represents an upper limit where a sufficiently large temperature gradient can be applied with low input thermal power.

0.6% ErAs:*n*-Type InGaAlAs

EDX spectroscopy was used to examine material compositions and therefore confirm material uniformity. EDX analyzes the elemental x-ray fingerprint emitted by the sample in response to being struck by electron beams. Figure 2 shows the elemental composition of individual points along the growth direction of the ErAs:InGaAlAs sample. The composition has less than 5% variation across the sample.

The TR coefficient was calibrated by placing the sample on top of a TE generator attached to a heat sink, as shown in Fig. 3. The temperature of the TE generator (and sample) was modulated using a sinusoidal current. A TR image of the sample was obtained in the usual manner, and simultaneously the temperature on the top of the sample was measured with a microthermocouple. A small thermocouple was used to ensure good thermal contact and fast response time. The TR amplitude was then divided by the amplitude of the temperature oscillation as determined by the thermocouple to give the TR coefficient κ. Such calibration of the TR coefficient of the ErAs:InGaAlAs TE element was performed, and a pixel average over the images was used to calculate $\Delta R/R$. A TR coefficient of $\kappa = 0.0005\ \mathrm{K}^{-1}$ was found using temperature modulations with amplitude between 0.5 K and 1.75 K.

After confirming the uniformity of the sample and determining the TR coefficient, the instrument was utilized to examine thermal conductivity. The sample was cleaved with a smooth surface, then cleaned and mounted between the measurement bar and the heating element. A 50-mHz sinusoidal current (3 A peak to peak) was injected into the TE heater, and phase-locked to the camera using two coupled function generators. Images were then taken using trigger frequency of 200 mHz. For each measurement, 20,000 averaging iterations were performed.

Different temperature gradients were created by the heating element with different applied current values, while the heat flux was measured by thermocouples. The temperature profiles across the sample are shown in Fig. 4. The images have been averaged in the transverse direction to improve the temperature resolution and obtain a thermal profile of the element along the direction of heat flux. The linear temperature profile indicates that the thermal conductivity remains constant across the sample during the long (24 h) MBE growth of the sample, showing that the thermal conductivity has less than 7% variation across the sample.

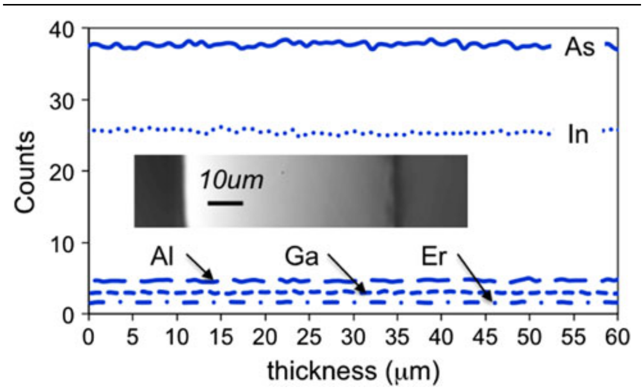

Fig. 2. X-ray diffraction of ErAs:InGaAlAs TE elements. The inset shows a photomicrograph of the cross-plane view.

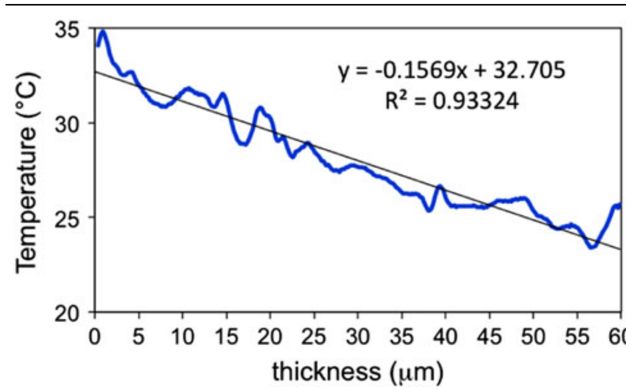

Fig. 4. Temperature profile of the ErAs:InGaAlAs TE element at modulation frequency of 50 mHz.

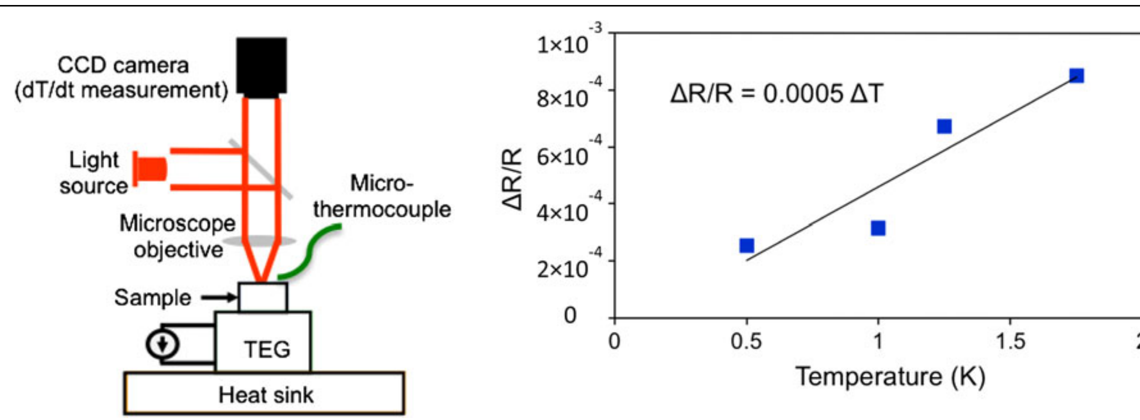

Fig. 3. Schematic diagram of apparatus for TR coefficient calibration and TR coefficient of ErAs:InGaAlAs.

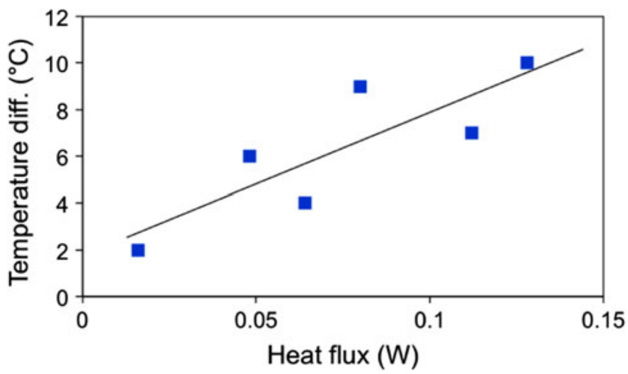

Fig. 5. Temperature difference across an ErAs:InGaAlAs TE element as a function of heat flux.

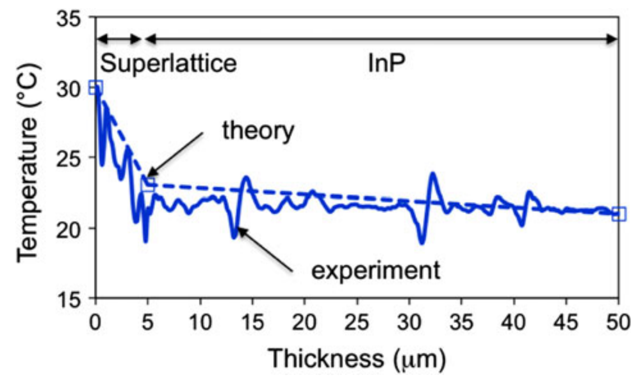

Fig. 6. Temperature profiles of ErAs:InGaAlAs on InP substrate.

From the temperature profile of the TE element, a thermal conductivity value of 2 W/mK was extracted using the heat flux information from the measurement bar and the one-dimensional (1D) steady-state heat conduction equation (1).

$$k = -\frac{Q}{A}\frac{\mathrm{d}t}{\mathrm{d}T}, \quad (1)$$

where Q is the heat flux, k is the thermal conductivity, A is the area, t is the thickness, and T is the temperature.

To further improve the measurement accuracy, different temperature gradients were created and therefore different heat fluxes measured across the sample. Linear curve fitting of temperature gradient versus heat flux is shown in Fig. 5. A thermal conductivity value of 2.3 W/mK was extracted from Eq. 2.

$$k = -\frac{t}{A}\frac{\mathrm{d}Q}{\mathrm{d}T}, \quad (2)$$

where Q is the heat flux, k is the thermal conductivity, A is the area, t is the thickness, and T is the temperature. The average thermal conductivity obtained with the 3-omega method was 2.5 W/mK.[13]

ErAs:*n*-Type InGaAlAs on InP Substrate

The instrument was also utilized to examine thermal conductivity variations across a sample consisting of 5 μm of ErAs:*n*-type InGaAlAs on a 45-μm InP substrate. A 100 mHz sinusoidal current (3 A peak to peak) was injected into the TE heater, and phase-locked to the camera using two coupled function generators. Images were then taken using trigger frequency of 400 mHz. For each measurement, 20,000 averaging iterations were performed.

The temperature profile across the sample is shown in Fig. 6. The image was averaged in the transverse direction to improve the temperature resolution and obtain a thermal profile of the element along the direction of heat flux. Two temperature slopes were observed, indicating two different thermal conductivities. Most of the temperature drop is across the ErAs:*n*-type InGaAlAs TE element, which shows it has lower thermal conductivity (1.9 W/mK) than the InP substrate (68 W/mK).

CONCLUSIONS

A micro-instrumentation utilizing the TR technique to characterize thermal conductivity and material uniformity is developed. By using this technique, we can achieve high spatial resolution for thermal conductivity and eliminate contributions from thermal contact resistance, but also eliminate the need for sample preparation other than cleaving. This method offers temperature resolution of 10 mK, spatial resolution of 200 nm, and thermal conductivity measurement with 0.01 ± 0.001 W/mK resolution. This method has been verified against the 3-omega technique, and the results of the two methods are in good agreement. Besides, a temperature profile of a bilayer sample is imaged with this technique. Two different slopes are observed, indicating different thermal conductivities.

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