Thin film ZT characterization using transient Harman technique

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Abstract

Thin-film thermoelectric materials offer great potential for improving the thermoelectric figure of merit ZT due to the freedom of tailoring the electron and heat transport. The characterization of these thin films is difficult because of the coexistence of the substrate, non-ideal contact, and asymmetric three-dimensional device structure. We have investigated theoretically and experimentally the transient Harman method for measuring the ZT of a thin film Si/SiGe superlattices on a silicon substrate. 3D electrothermal simulations allow us to identify the contribution of the thin film and the substrate to the transient response. On the measurement side, ringing at short times and noise can be significantly improved by using high-speed packages and electrical impedance matching. The Joule heating contribution to the thermoelectric EMF is separated from the Peltier one by the bipolar measurement. The parasitic non-ideal effects of contacts and substrate can be removed by variable thickness superlattice method.

Introduction

Low-dimensional thin film thermoelectric materials have demonstrated a large improvement in the thermoelectric figure of merit ZT [1]. This is mainly due to the increased phonon scattering of the heterostructures and therefore the reduced thermal conductivity. Another benefit comes from the asymmetric narrow band of the density of states of the quantum confined system. However, it is a great challenge to characterize these thin film structures because of the micron level thickness and nanoscale quantum size. The parasitic substrate and non-ideal contact and packaging make it very difficult to extract the small and fast signal of the thin film itself. There have been some measurements of the electrical conductivity, thermal conductivity and the Seebeck coefficient in tackling this difficulty [2-4]. Transient Harman method is a convenient way to get the ZT without any micro scale temperature measurement [5]. In this paper, we investigate the effectiveness of this method on extracting the ZT of the SiGe superlattice. A theoretical model is developed to treat the non-symmetric thermal path and non-ideal contact. The 3D finite element simulation shows that the parasitic substrate and contact effects can be separated by the variable thickness method.

Theoretical Model and Simulation

In a typical transient Harman method of measurement of the ZT of the thermoelectric materials, a DC current is first applied to the sample. The Peltier effect results in a temperature difference across the thermoelectric material, which in turn generates a Seebeck voltage against the flow of the current. Thus the total voltage across the sample is the sum of electrical voltage of Ohm's law and Seebeck voltage due to Peltier effect, as shown in Fig. 1. When the current is turned off, the electrical voltage V_R disappears instantaneously, while the Seebeck voltage V_S drops slowly because of the heat dissipation and heat capacity of the materials. This provides a convenient way of measuring the ZT since it is the ratio of Seebeck voltage and electrical voltage.

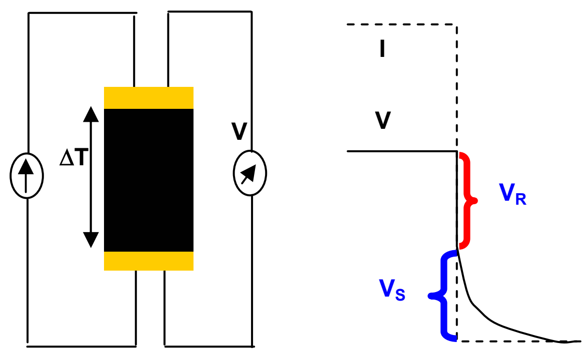

Fig. 1. Schematic of the transient Harman method.

However, for a real thin film device as shown in Fig. 2, there are a lot of parasitic substrate resistance and non-ideal contact resistance, so that the electrical voltage is not that across the thin film only. Furthermore, not only Peltier effect, but also Joule heating (the red arrows indicate the current flow) makes up of the Seebeck voltage, because the device structure and heat sinking are non-symmetric and the Joule heating is distributed widely and non-uniformly.

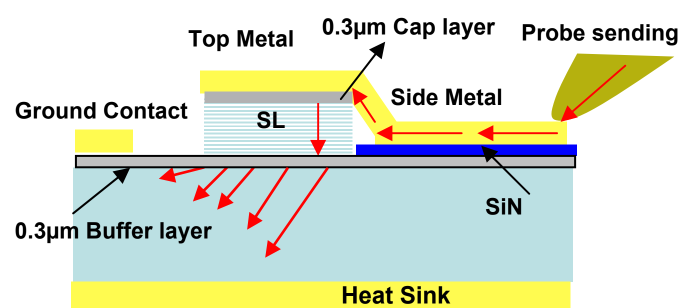

Fig. 2. The structure of a SiGe superlattice micro refrigerator device.

To check the contributions of Joule heating and Peltier effect to temperature distribution and gradient, the heat equation can be written as

$$\rho c \frac{\partial T(\vec{r},t)}{\partial t} = k\nabla^2 T \pm Q_{sp}(\vec{r},t) + Q_{sJ}(\vec{r},t).$$

where ρ, c and k are the mass density, capacity, and thermal conductivity, respectively. On the right side, the first term is heat conduction, the second is Peltier effect, and the third is Joule heating.

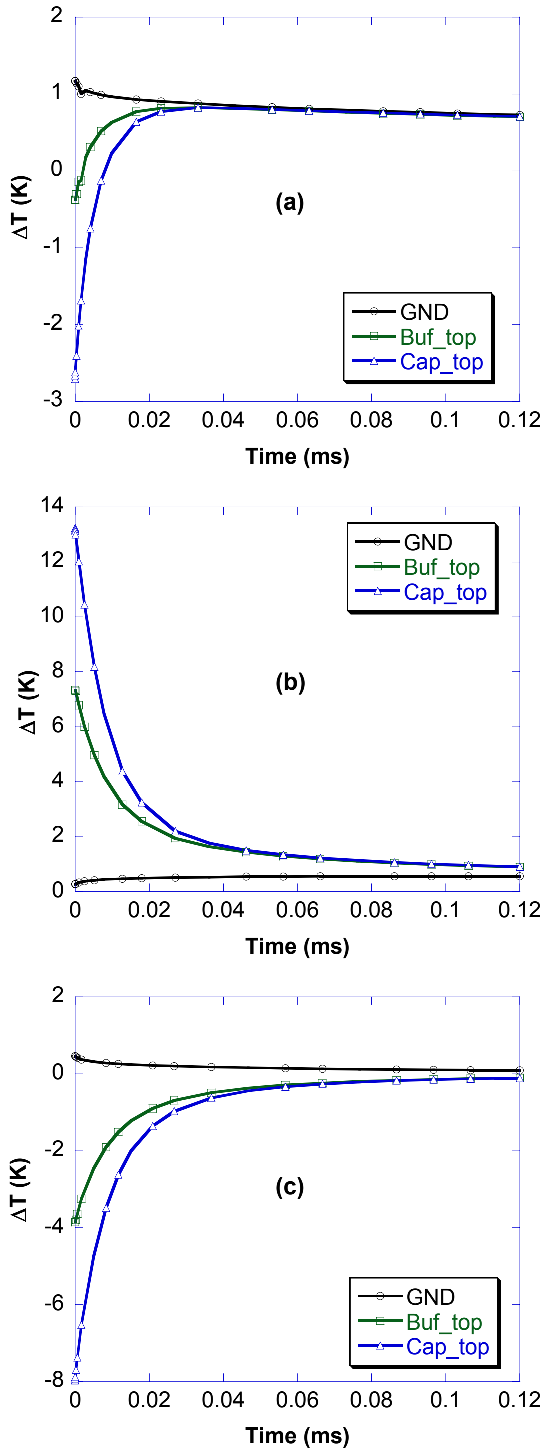

Fig. 3. Finite element simulation of the transient temperature response of a micro cooler to a 400 mA current when (a) both Joule heating and Peltier effect are included for direct bias, (b) both Joule heating and Peltier effect are included for reverse bias, and (c) only Peltier effect is included for direct bias.

The sign of the Peltier effect is determined by the current direction and Seebeck coefficient distribution. And the Peltier cooling or heating is only produced at the interface of hetero materials when there is a change of Seebeck coefficient. The Joule heating is distributed in the device and it is difficult to account for its effect in a 3D geometry. Because the heat equation is linear and the Joule part is independent of the current direction, it can be removed by a subtraction of the reverse current measurement from the direct current one with the same amplitude. The Peltier effect just changes the sign so that the contribution to the Seebeck voltage from the Peltier effect can be separated from the contribution due to Joule heating.

We used the ANSYS finite element tool to simulate the temperature distribution and transient response of a SiGe superlattice micro cooler. For a device mesa measured $50\mu m \times 50\mu m$ and 3 microns thick thin film, when a 400 mA current is applied, the temperature transient responses of the direct and reverse current biasing including both Joule heating and Peltier effect are shown in Fig. 3 (a) and (b), respectively. The response of pure Peltier cooling effect is shown in (c), which is equal to the subtraction of (b) from (a) divided by a factor of 2. The monitored temperature positions are at ground, buffer layer top, and cap layer top, respectively.

However, after the removal of Joule heating contribution to Seebeck voltage, the Seebeck voltage across the substrate (which is not metal) is still included in the total as that across the superlattice thin film. And the measured electrical voltage consists of those across the thin film, substrate and any contact resistance. One good way to remove these parasitic voltages is by variable thickness method, assuming different thin film thickness samples fabricated by the same processing have the same material and contact properties. This method is obviously effective for electrical voltage since it is linearly dependent on the thickness of the thin film,

$$V_E = I\frac{l}{\sigma A} + IR_{E-sub}$$

where I, l, σ, A, $R_{E\text{-}sub}$ are the current, thickness of the thin film, electrical conductivity, cross area, and equivalent resistance of the substrate, respectively. However, the thermal circuit is complicated (as shown in Fig. 4) and there may be some heat leakage from the contact leads to either the substrate or the air.

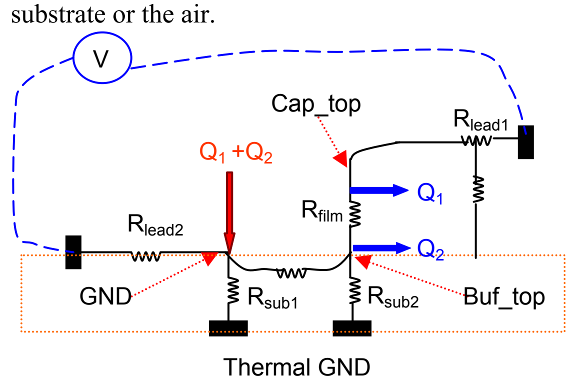

Fig. 4. The thermal circuit of the micro cooler device.

The thermal resistance is not symmetric relative to the position of thin film. If the electrical current is constant, only when the heat leakage from the top and side is much smaller than the heat flow through the thin film and substrate, the temperature across the substrate is constant and that across thin film is proportional to its thickness for Peltier effect.

$$\Delta T_{film} = ITS_{film} \frac{l}{kA} \propto l$$

This in turn makes the total Seebeck voltage of Peltier effect is linear to the thickness of thin film.

$$V_{sp} = ITS_{film}^2 \frac{l}{kA} + V_{s-sub}$$

Then the ZT of the material can be calculated by

$$ZT = \frac{S^2\sigma}{k} T = \frac{\partial V_{sp} / \partial l}{\partial V_E / \partial l}$$

In Fig. 5, we plot the simulated temperatures at the three monitored points of different thin film thicknesses for Peltier effect. It is shown that the temperatures at the contact ground and buffer top are almost constant, while that at the cap top is linear to the thickness. This means the temperature difference across the thin film is proportional to its thickness, if a real device structure and packaging is used in the simulation.

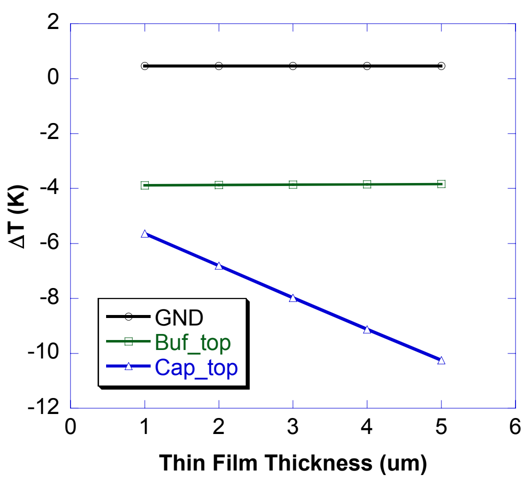

Fig. 5. Temperature for different thin film thicknesses at steady state when only Peltier effect is included.

There are several reasons making the transient ZT measurement of thin film a challenge task. First, since the thin films are several microns thick, the heat diffusion time is at micro second scale. Second, the resistance of the thin film device is typically below 1 Ohm, the impedance matching to the regular 50 Ohms transmission line is requested in a high speed measurement. In the transient measurement, usually the initial data points exhibit ringing due to impedance mismatch and should be excluded. A better estimation of the voltage at the current pulse falling edge can be achieved by curve fitting of the remaining "good" data points and extending back to the falling edge. We prove this argument by a comparison of the whole finite element transient simulation results of the temperature with those curve fittings (sum of three exponential decays) on the data points starting about 1.6 micro seconds after the current falling edge, shown in Fig. 6. The real values are very close to those predicted values at t=0 by curve fitting of the data points between 1.6 and 20 micro seconds for the substrate, and substrate with top thin film thickness of 1, 3 and 5 micrometers, respectively.

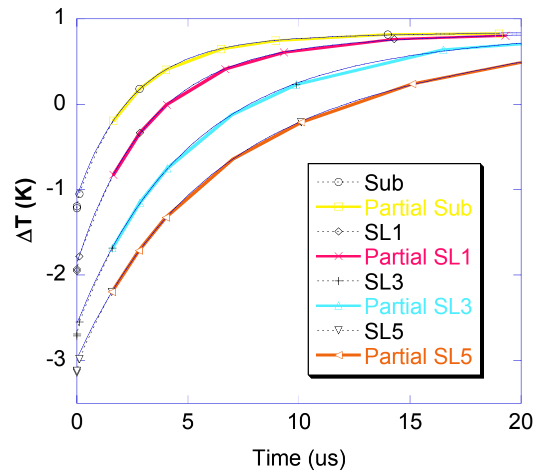

Fig. 6. The comparison of the real data and curve fitting.

Conclusions

We demonstrate by finite element simulation of a real micro cooler device that transient measurement is an effective method to obtain the ZT value of a thin film thermoelectric device. The subtraction of reverse current result from that of direct biasing can remove the contribution of Joule heating to Seebeck voltage. The variable thickness method can separate the electrical voltage and Seebeck voltage of parasitic substrate and contacts if the device processing technique gives similar results for different devices and the heat leakage through the top and side lead is much smaller than the conduction through the thin film. Curve fitting can provide much better reading of the Seebeck voltage at the falling edge of the current.

Acknowledgments

This work is supported by the Office of Naval Research (ONR) - Multidisciplinary University Research Initiative (MURI) Grant, Thermionic Energy Conversion Center.

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