Characterization of Materials with Epitaxially Embedded Nanoinclusions for Thermoelectric Applications

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Abstract:

The engineering of thermoelectric materials for better electrical and thermal transport properties is an essential part of increasing the efficiency of thermoelectric modules and generators. In this work, we report on the implementation of the third harmonic (3ω) method for measuring the thermal conductivity of thin films. At the time of this writing, proof-of-concept tests for measuring thermal conductivity of silicon dioxide (SiO_2) thin films on Si and aluminum oxide (Al_2O_3) substrates were successfully completed. Additionally, the effect of doping III-V semiconductor compounds with rare-earth semimetal nanoparticles is reported through comparison of simulation results to published experimental data.

Background:

More than half of all energy generated and imported into the United States is wasted [1]. The vast majority of this wasted energy takes the form of heat. Recovering this waste heat for the improvement of power system efficiencies has become a major research problem in the field of thermoelectric materials [2-4].

Recent studies have demonstrated the benefits of tailoring thermoelectric materials on the nanoscale [4-6]. Comparisons of these materials are made via a dimensionless figure of merit, ZT, which includes the following parameters related by Equation 1: Seebeck coefficient (S), electrical conductivity (σ), thermal conductivity (κ), and absolute heat source temperature (T).

$$ZT = \frac{S^2\sigma}{\kappa}T$$

Equation 1

To become commercially useful, ZT values of 1.5-2.0 are desired [4].

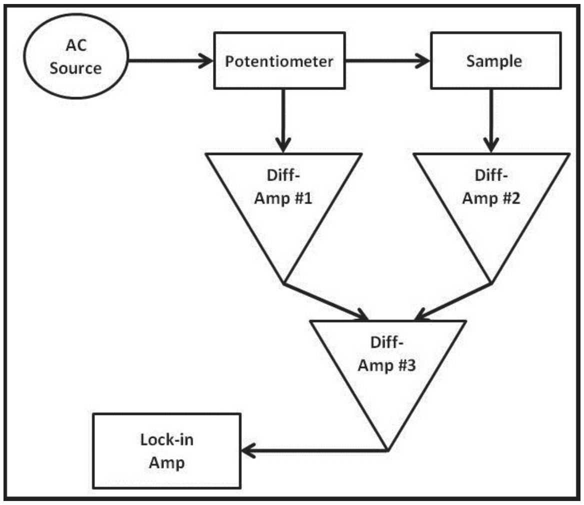

Figure 1: Functional block diagram of 3ω apparatus.

Experimental Methods:

All measurements and simulations in this report assume room temperature and pressure.

Thermal conductivity was measured according to the 3ω method [7, 8]. This method was originally proposed by Cahill, et al., for thick samples, and was later modified for measurement on thin-films. The experimental apparatus is outlined in Figure 1. The potentiometer was tuned such that the output of diff-amp #3 approached zero, eliminating the 1ω signal from the source. Only the sample contributed a substantial 3ω voltage, which was read by the lock-in amp. This 3ω voltage was proportional to the thermal conductivity.

To improve the ratio of the third harmonic with respect to the first, a signal conditioning circuit was built, the design of which was closely based on available circuits in the literature.

The Seebeck coefficient of materials was determined by measuring the voltage across a sample, upon which was applied a varying temperature gradient. Thermocouples and voltmeters were used to capture the Seebeck coefficient, which is defined as the ratio of open circuit voltage to the temperature gradient. A MatLab© routine was used to automate the temperature sweep and data collection.

Carrier concentration, n, was determined via the Hall effect, and electrical conductivity, σ, was determined via the van der Pauw method. Hall carrier mobility, μ, was found through the relationship $\sigma = nq\mu$, where q is the elementary charge.

MatLab© code created by A.T. Ramu [9] for calculating Seebeck coefficient of III-V compounds was calibrated via comparing simulation results of silicon-doped gallium arsenide, as a function of carrier concentration, with measurement results. The ultimate purpose of the simulation work was to characterize the improvement in the Seebeck coefficient of indium gallium aluminum arsenide (InGaAlAs) by doping with erbium arsenide (ErAs) nanoparticles [6], compared to a control sample of InGaAlAs doped with Si.

Results and Conclusion:

To confirm the proper implementation of the 3ω experimental apparatus, a sample from UC Berkeley with known thermal conductivity was measured and the results compared to our measurements. As evident from Figure 2, the measurements agree within 3% of each other.

The results of the simulations are presented in Figure 3. The Seebeck coefficient of 0.6%ErAs:InGaAlAs was found to be from 15% to 35% greater than the control Si:InGaAlAs. The carrier concentration of the simulated Si:InGaAlAs was fixed at 1.9e18cm^{-3} since optimum thermoelectric properties of Si:InGaAlAs are found at this concentration.

In conclusion, we have established the 3ω method at UCSB and illustrated the benefits of ErAs doping. Future work of immediate relevance to this report consists of using the 3ω apparatus for measurement of thermal conductivity of the III-V semiconductor compounds doped with rare-earth nanoparticles and expanding the simulation programs, e.g., enabling alloy scattering for InGaAlAs, adding more exotic material systems.

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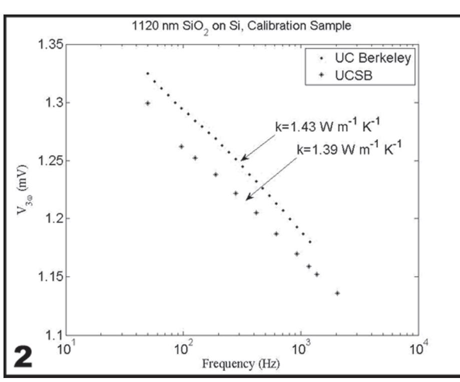

Figure 2: Verification of implementation of 3ω method.

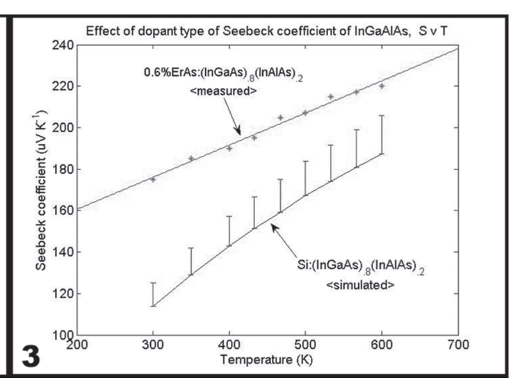

Figure 3: Error in the simulated Seebeck coefficient of Si:InGaAlAs is based on variance from measurements and neglect of alloy scattering.