

Thermal Conductivity Measurements of ErAs:InGaAlAs for Thermoelectric Applications

S.L. Singer¹, W. Kim¹, A. Majumdar^{1*}
J.M.O. Zide², A.C. Gossard²

¹University of California, Department of Mechanical Engineering
Etcheverry Hall, Berkeley, CA 94710, U.S.A.

²University of California, Department of Materials
Santa Barbara, CA 93106, U.S.A.

Abstract

The efficiency of thermoelectric materials are typically described using the thermoelectric figure of merit, ZT , where $ZT=S^2\sigma T/k$. S , σ , and k are the Seebeck coefficient, electrical conductivity, and thermal conductivity respectively. A high power factor ($S^2\sigma$) and low thermal conductivity (k) are essential for efficient operation of thermoelectric devices. We incorporated ErAs nanoparticles in an InGaAlAs (ErAs:InGaAlAs) matrix epitaxially to lower the thermal conductivity yet maintain the power factor of InGaAlAs. We show that the thermal conductivity of 0.3% ErAs:InGaAlAs does not differ much from that of 0.3% ErAs:InGaAs. However, a reduction in thermal conductivity for 3% ErAs:InGaAlAs emphasizes the importance of using nanometer size particles in addition to alloy substitutions in a matrix to reduce thermal conductivity.

Keywords: Thermoelectric, Thermal Conductivity, InGaAs, InGaAlAs

1 - INTRODUCTION

The efficiency of thermoelectric materials are typically described using the thermoelectric figure of merit, ZT , as described by Eq. (1).

$$ZT=S^2\sigma T/k \quad (1)$$

S , σ , and k are the Seebeck coefficient, electrical conductivity, and thermal conductivity respectively. A high power factor ($S^2\sigma$) and low thermal conductivity (k) are essential for efficient operation of thermoelectric devices. Recent increases in ZT by using nanostructures mainly come from the thermal conductivity reduction [1]. By embedding nanoparticles in the alloy matrix, the thermal conductivity can be reduced even at high temperatures without sacrificing the power factor [2].

2 - THERMOELECTRIC MATERIALS

The materials studied are (i) 0.3% ErAs randomly distributed in $\text{In}_{0.53}\text{Ga}_{0.28}\text{Al}_{0.19}\text{As}$ (structure A), (ii) 0.3% ErAs randomly distributed in $\text{In}_{0.52}\text{Ga}_{0.09}\text{Al}_{0.39}\text{As}$ (structure B), and (iii) 3% ErAs randomly distributed in $\text{In}_{0.53}\text{Ga}_{0.28}\text{Al}_{0.19}\text{As}$ (structure C). Self-assembled ErAs nanoparticles are epitaxially embedded in InGaAs which is grown by molecular beam epitaxy (MBE). Both 0.3%-ErAs:InGaAlAs samples (structure A and B) are 2 μm and grown on a semi-insulating InP substrate with a buffer layer of 100nm InAlAs as shown in Fig.1a. The 3% ErAs:InGaAlAs sample (structure C) is 1 μm and grown on a semi-insulating InP substrate with a buffer layer of 50nm on InAlAs as shown in Fig.1b.

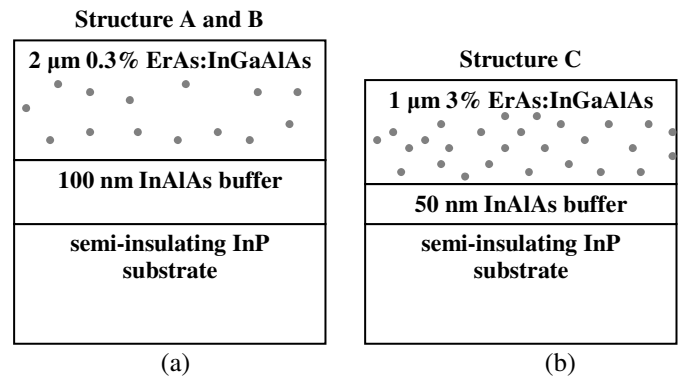


Figure 1 – Sample structure of ErAs:InGaAlAs. (a) Samples with 0.3% ErAs randomly distributed in InGaAlAs on a 100 nm InAlAs buffer layer on a semi-insulating InP substrate; structure A and B. (b) Samples with 3% ErAs randomly distributed in InGaAlAs on a 50 nm InAlAs buffer layer on a semi-insulating InP substrate; structure C.

As a reference, we will compare the thermal conductivity of the ErAs:InGaAlAs samples to $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ (InGaAs), 0.3% ErAs:InGaAs [2], and 3% ErAs:InGaAs. The growth method can be found in the literature [3]. The 3% ErAs:InGaAs film was a 1 μm layer grown on a semi-insulating InP substrate with a buffer layer of 50 nm InAlAs.

* Contact author: Tel. 510-643-8199, email: majumdar@me.berkeley.edu

3 - PREVIOUS WORK OF ErAs:InGaAs SYSTEMS

The scattering of phonons in an alloy is due to a difference in mass and/or bond stiffness dominated by atomic substitutions. The atomic size regime follows that of Rayleigh scattering which is responsible for short wavelength scattering. The thermal conductivity of an alloy has a minimum thermal conductivity known as the alloy limit. Kim et al. [2] showed that a further reduction in thermal conductivity below the alloy limit is possible due to additional scattering of mid to long wavelength phonons brought on by incorporating nanoparticles approximately 2-5 nm in diameter to an alloy. They recorded a reduction in thermal conductivity by nearly a factor of 2 for 0.3% ErAs:InGaAs when compared with the alloy limit of InGaAs at 300K. The thermal conductivity of both In_{0.53}Ga_{0.47}As and 0.3% ErAs:InGaAs will be shown as a reference [2].

4 - MEASUREMENT TECHNIQUE

Cross plane thermal conductivity measurements were made using the 3ω technique as described by Cahill [4]. We deposited Pt (~ 400 nm) with Cr (~ 40 nm) as an adhesion layer on SiO₂ (~ 0.18 μ m) to insulate the 30 μ m thick heater/thermometer pattern from the film. Thermal conductivity measurements were performed in a cryostat and we report results for the temperature range 100 K – 550 K.

5 - RESULTS

The thermal conductivity of 0.3% ErAs:InGaAs is clearly much lower than that of InGaAs as shown in Fig. 2. The reduction in thermal conductivity was evident over the entire measured temperature range, but most prominent from 100K – 300K. The thermal conductivity of 0.3% ErAs:InGaAlAs samples (structure A and B) are similar to that of 0.3% ErAs:InGaAs. Atomic incorporations, such as replacing In or Ga with Al to create alloys, usually affect high frequency phonons. Atomic substitutions in an alloy similarly prefer to scatter high frequency phonons, which does not produce a significant difference in thermal conductivity.

The thermal conductivity of 3% ErAs:InGaAlAs (structure C) is reduced by more than a factor of 2 below that of 0.3% ErAs:InGaAlAs. The atomic substitutions of InAlAs for InGaAs do not significantly reduce thermal conductivity for the case of 0.3% ErAs. However, there are some differences between the 3% ErAs:InGaAs and 3% ErAs:InGaAlAs samples. More experimental studies are ongoing to understand this behavior. For the same percentage of InAlAs in the ErAs:InGaAlAs samples, increasing the ErAs concentration (not necessarily the size of ErAs nanoparticles) to 3% significantly reduced the thermal conductivity. This observation demonstrates the importance of phonon scattering by the ErAs nanoparticles.

6 - CONCLUSIONS

Incorporating 0.3% of ErAs nanoparticles into InGaAlAs reduces the thermal conductivity by nearly a factor of two below InGaAs at 300K. The thermal conductivity of 0.3% ErAs:InGaAlAs with varying percentages of InAlAs does not

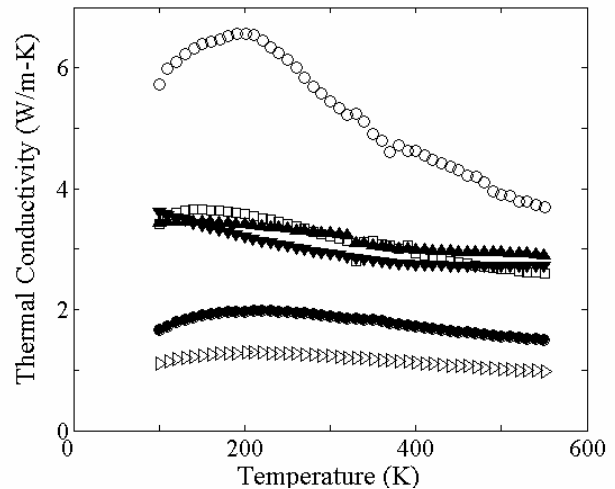


Figure 2 – Thermal Conductivity of InGaAs (open circles), randomly distributed 0.3%-ErAs:InGaAs (open squares), 0.3%-ErAs:In_{0.53}Ga_{0.28}Al_{0.19}As (structure A, closed triangle down), 0.3%-ErAs:In_{0.52}Ga_{0.09}Al_{0.39}As (structure B, closed triangle up), 3% ErAs:InGaAs (closed circles), and 3%-ErAs:In_{0.53}Ga_{0.28}Al_{0.19}As (structure C, open right triangles)

vary much compared to 0.3% ErAs:InGaAs. This suggests that since high frequency phonons are already scattered by the alloy structure, replacing one high frequency scattering mechanism with another does not produce much difference.

Increasing the concentration of ErAs nanoparticles to 3% in InGaAlAs reduces the thermal conductivity by more than a factor of 2 below that of 0.3% ErAs in InGaAlAs. This suggests that increasing the concentration of nanoparticles indeed play an important role in thermal conductivity reduction.

ACKNOWLEDGMENTS

This work was supported by the Office of Naval Research (ONR) Multidisciplinary University Research Initiative (MURI) Grant. S.L.Singer thanks the National Science Foundation (NSF) for a personal fellowship

REFERENCES

- [1] Majumdar, A., "Thermoelectricity in Semiconductor Nanostructures," *Science*, Vol. 303, pp. 777-778, February 2004.
- [2] Kim, W., et al., "Thermal Conductivity Reduction and Thermoelectric Figure of Merit Increase by Embedding Nanoparticles in Crystalline Semiconductors," *Phys. Rev. Lett.*, Vol. 96, (045901), 2006.
- [3] Zide, J.M.O., et al., "Thermoelectric power factor in semiconductors with buried epitaxial semimetallic nanoparticles," *Appl. Phys. Lett.* Vol. 87, (112102), 2005.
- [4] Cahill, D.G., et al., "Thermal conductivity measurement from 30 to 750 K: the 3ω method," *Rev. Sci. Instrum.* Vol. 61, pp. 802-808, 1990.