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Thermoelectric Generators of Sequentially Deposited Si/Si+Ge Nano-layered Superlattices

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Abstract

Effective thermoelectric materials have a low thermal conductivity and a high electrical conductivity. The performance of the thermoelectric materials and devices is shown by a dimensionless figure of merit, $ZT = S^2\sigma/ K_{TC}$, σ is the electrical conductivity T/ K_{TC}, where S is the Seebeck coefficient, T is the absolute temperature and K_{TC} is the thermal conductivity. In this study we have prepared the thermoelectric generator device of Si/Si+Ge multi-layer superlattice films using electron beam physical vapor deposition (EB-PVD). 5 MeV Si ion bombardment was performed in the multi-layer superlattice thin films to decrease the cross plane thermal conductivity, increase the cross plane Seebeck coefficient and cross plane electrical conductivity.

Keywords: Ion bombardment, thermoelectric properties, multi-nanolayers, Figure of merit.
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1. Introduction

The performance of a thermoelectric device is quantified by the dimensionless figure of merit $ZT=S^2\sigma T/K_{TC}$. Our aim is to obtain high ZT values by increasing the Seebeck coefficient S and the electrical conductivity σ, and reducing the thermal conductivity KK_{TC} by bombarding the structure with MeV Si ions. Ion bombardment induces the formation of quantum dots of Si and Ge. In addition to, the quantum well confinement of phonon transmission due to Bragg reflection at lattice interfaces [1,2] the defects and disorder in the lattice caused by ion bombardment and the grain boundaries of these nanoscale clusters increase phonon scattering and increase the chance of an inelastic interaction and phonon annihilation. All these effects inhibit heat transport perpendicular to the layer planes [4–7]. Thus, cross plane thermal conductivity will decrease. These quantum dot layers also increase the Seebeck coefficient and electric conductivity owing to the increase of the electronic density of states produced by the one dimensional periodic potential. We have already studied the improvement of thermoelectric properties for 10–

50 nm multilayers [8]. In this study we report on the growth of Si+Ge multi-layer super-lattice films using co-electron beam physical vapor deposition (EB-PVD). Si and Ge materials were placed in separate electron guns and the shutters were manipulated to deposit the desired materials at the desired thickness. The deposition was followed by a 5 MeV Si ion bombardment at various fluences.

2. Sample preparation and characterization

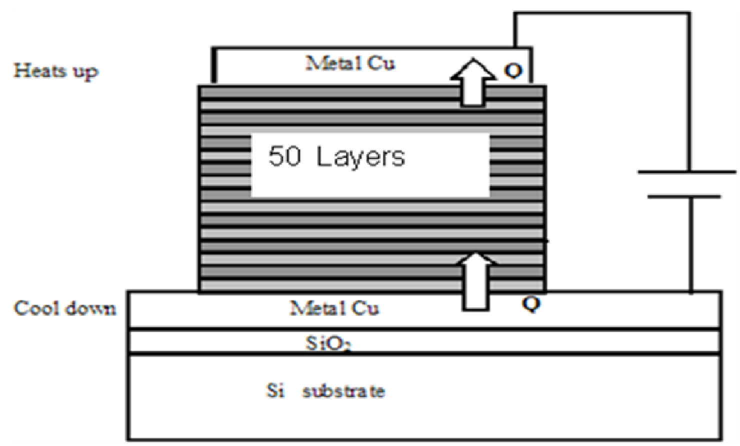

Fig.1. Diagram of electrical conductivity measurements.

Fig. 1 shows the multilayer preparation geometry and the diagram for the electrical conductivity and Seebeck measurement set-up. Si/Si+Ge multilayer thin film thermoelectric (TE) devices were made at the AAMU Center for Irradiation of Materials. These thin films constitute a periodic quantum well structure consisting of 50 alternating layers of total thickness of 300nm. The multilayers were prepared by electron beam physical vapor deposition (EB-PVD). The process pressure 2×10^{-5} Torr was maintained throughout the deposition. The multilayer films were sequentially deposited on a Si substrate that was coated with a SiO_2 insulation layer and a metal (Cu) contact layer to form a multilayer. A quartz crystal monitor (QCM) was used to monitor deposition rate and final thickness. For each Si layer the relative rate of deposition was 10 Hz/s from a single e-beam evaporator. The thin film was grown on a carbon substrate for Rutherford backscattering spectroscopy (RBS) analysis. Post 5 MeV Si ion beam process was performed using the AAMU Pelletron accelerator. (SRIM-2008) Stopping Range Ions in

Matter simulation shows that 5 MeV Si ions pass through the multilayer film and terminate deep in the substrate.

The electrical conductivity was measured by the 4-probe contact system and the thermal conductivity was measured by the 3ω technique. The electrical conductivity, thermal conductivity and Seebeck coefficient measurements have been performed at room temperature. In order to make nano clusters in the layers, 5 MeV Si ion bombardments were performed with the Pelletron ion beam accelerator at the Alabama A&M University Center for Irradiation of Materials (AAMU-CIM).

The energy of the bombarding Si ions was chosen by the SRIM-2008 simulation software (SRIM-2008). The fluences used for the bombardment were $1x10^{12} ions/cm^2$, $5x10^{12} ions/cm^2$ and $1x10^{13} ions/cm^2$. Rutherford Backscattering Spectrometry (RBS) was performed using 2.1 MeV He^+ ions with the particle detector placed at 170 degrees from the incident beam to monitor the film thickness and stoichiometry before and after 5 MeV Si ion bombardments [9].

3. Thermoelectric measurements and results

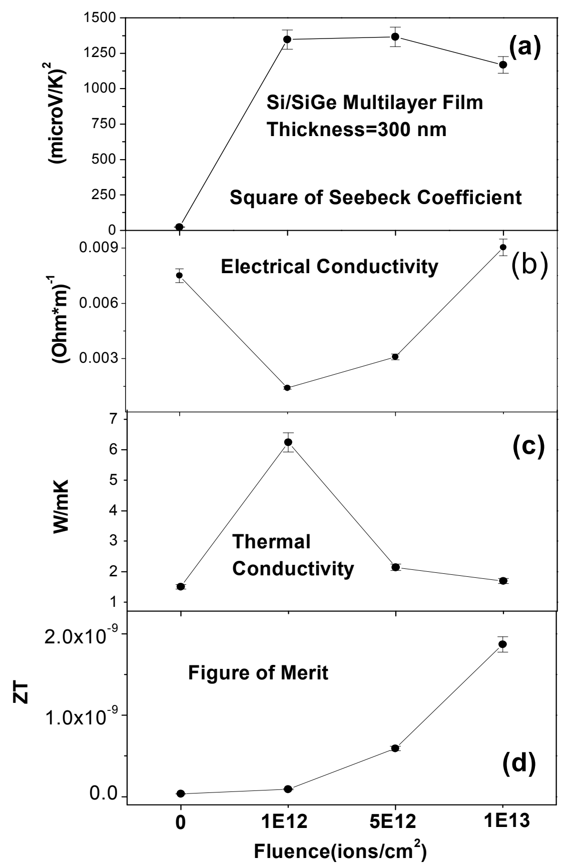

Fig. 2. Results of Electrical and Thermoelectric measurements

We used the 3ω technique to measure the cross plane thermal conductivity of the thin film samples. The experimental setup has been previously described [3,4,8]. A narrow Ag strip is deposited onto the films providing a heater with a resistance value of about 200 Ω. Originally we got negative Seebeck values for this sample. The negative value of the observed Seebeck coefficient indicates that our samples are n type. For cross plane Seebeck coefficient measurements, the samples are deposited between two thermally and electrically conductive metal (Cu) layers as shown in Fig. 1. The Square of Seebeck coefficients of the multilayer samples before and after 5 MeV Si ion bombardments are compared in Fig. 2a. The electrical conductivities of Si/ Si + Ge multilayer thin films before and after bombardment by 5 MeV Si ions are shown in Fig. 2b. We used a digital electronic bridge with four probe contact system to measure the cross plane electrical conductivity of the thin film samples. We assumed that the Schottky junction barrier between Cu and the semiconductors are negligible and that the resistance of the Cu surface electrodes is negligible. Fig. 2c indicates that cross plane thermal conductivity of Si/ Si + Ge multilayer thin films before and after bombardment by 5 MeV Si ions. As seen from fig. 2c, the thermal conductivity value decreases with increasing ion fluence except for the value at the fluence of $1x10^{12} ions/cm^2$. Finally, we have calculated the figure of merit ZT from the definition $ZT=S^2\sigma T/K$ before and after bombardment by 5 MeV Si ions and the results were shown in fig.2d.

4. Discussion and conclusion

In this study we deposited 50 layers as compared to previous studies where we have done on the order of 70 layers. The results here show a slight difference in the seebeck coefficient. This can be attributed to variations in processing parameter leading to variations in the layers of the materials. The increased number of charge carriers due to 5 MeV Si ion bombardment is most dominate feature of this technique for the increasing the electrical conductivity and the seebeck coefficient. The decrease in the thermal conductivity can be related to both the multilayer interface and Si bombardment. We suspect that an increase of thermal conductivity can be accomplished with increasing the homogeneity of the multilayer. By providing more defined interfaces, thus increasing the phonon scattering. Naturally , as the fluence of the ion bombardment increases the more damage is caused in the layers and more importantly the interfaces of the multilayers. We have yet to confirm the presence of nano cluster by microstructure analysis methods such as transmission electron microscopy (TEM). The optimum fluence will yield the greatest number of carriers and phonon scattering sites while minimizing the damage due to post ion beam bombardment.

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References

[1] Xiaofeng Fan, Electron. Lett. 37 (2001) 126.
[2] Xiaofeng Fan, Gehong Zeng, Appl. Phys. Lett. 78 (2001) 1580.
[3] S.M. Lee, D.G. Cahill, Appl. Phys. Lett. 70 (1997) 2957.
[4] D.G. Cahill, M. Katiyar, Phys. Rev., B 50 (1994) 6077.
[5] Rama Venkatasubramanian, Phys. Rev., B 61 (2000) 3091.
[6] J.L. Liu, Phys. Rev., B 67 (2003) 4781.
[7] David G. Cahill, Rev. Sci. Instrum. 61 (1990) 802.
[8] B. Zheng, S. Budak, C. Muntele, Z. Xiao, C. Celaschi, I. Muntele, B. Chhay,
[9]R.L. Zimmerman, L.R. Holland, D. Ila, Materials in Extreme Environments, Materials Research Society, vol. 929, 2006, p. 81.
[10] L.R. Doolittle, M.O. Thompson, RUMP, Computer Graphics Service (2002).
[11]S. Budak, S. Guner, R. Minamisawa and D. Ila, Surface and Coatings Technology, Vol. 203, issues 17-18,2009,P. 2479
[12] B. Zheng, Z. Xiao, B. Chhay, R. Zimmerman, D. Ila , Nuclear Instr. And Methods in Physics Research B 266 (2008) 73-78