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Improvement on thermoelectric properties of multilayered $Si_{1-x}Ge_x/Si$ by ion beam bombardment

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Abstract

We made n-type nano-scale thin film thermoelectric (TE) devices that consist of multiple periodic layers of $Si_{1-x}Ge_x/Si$. The period is about 10 nm. The structure was modified by 5 MeV Si ion bombardment that formed a nano-scale cluster structure. In addition to the effect of confinement of the phonon transmission, formation of nanoclusters by the ionization energy of incident MeV Si ions further increases the scattering of phonons, increasing the chance of inelastic interaction of phonons, resulting in more annihilation of phonons. This limits phonon mean free path. Phonons are absorbed and dissipated along the layers rather than in the direction perpendicular to the layer interfaces, therefore cross plane thermal conductivity is reduced. The increase of the density of electronic states due to the formation of nanocluster minibands increases the cross plane Seebeck coefficient and increases the cross plane electric conductivity of the film. Eventually, the thermoelectric figure of merit of the TE film increases.

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Keywords: Thermoelectric multilayer superlattice; Ion bombardment; Rutherford backscattering spectroscopy; 3ω measurement of thin film thermal conductivity

1. Introduction

Semiconductor thermoelectric devices are widely used as the cooling device for integrated circuits and the electrical power generator device because of their higher efficiency, smaller volume and higher power density. While Si and Ge can be monolithically integrated with Si-based microelectronic device. And thin film multilayer superlattice thermoelectric device with quantum well structure has a lower thermal conductivity, In order to further decrease phonon transmission and meanwhile increase Seebeck coefficient and electric conductivity of thin film, in this research, MeV Si ion bombardment is used to modify the film to form nanoclusters among the superlattice. The phonon confinement effect is enhanced by ion bombardment that forms nanoclusters. The defects and disorder in the lattice caused by bombardment and the grain boundaries of the nano-scale clusters increase phonon scattering, increasing the probability of inelastic interactions of phonons and annihilation of phonons. This limits phonon mean free path. The existence of these nanoclusters enhances the horizontal dissipation and absorption of phonons along the superlattice interface rather than perpendicular to superlattice. The cross plane thermal conductivity of an ion bombardment thin film decreases. The cross plane electric conductivity and the cross plane Seebeck coefficient of the ion bombarded thin film also increases due to the increased density of electronic states. Hence, the thermoelectric figure of merit of the film increases. Si and Ge are very commonplace materials used in thermoelectric device, therefore study on ion beam modified

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$Si_{1-x}Ge_x$/Si superlattice is particularly significant for testing the effect of ion beam modification on thermoelectric device.

The actual thermoelectric device consists of multiple n type and p type couples, while in this research, we just study one section (n type or p type) individually. As shown in Fig. 1, the Si/$Si_{1-x}Ge_x$ multilayer thin film thermoelectric (TE) devices prepared at CIM are periodic structures consisting of tens to hundreds of alternating layers, each with different band gap (Si: 1.12 eV, $S_{1-x}Ge_x$: 0.8–0.9 eV). Each multilayer film is deposited between two metal layers. Due to the Peltier effect between the semiconductor film and the metal, an electric current causes heat to be absorbed at the negative terminal and released at the positive terminal. The performance of a thermoelectric device can be quantified by the dimensionless figure of merit $ZT = S^2\sigma T/k$ [1]. Our aim is to obtain a high ZT value by increasing cross plane Seebeck coefficient S, cross plane electrical conductivity σ and reducing cross plane thermal conductivity k by bombarding a multilayer structure with MeV Si ions. Ion bombardment induces the formation of nano-scale clusters. In addition to the Bragg reflection at the layer interfaces [1–3], the defects and disorder in the lattice caused by ion bombardment and the boundaries of these nano-scale clusters formed by bombardment increase phonon scattering, increasing the chance of an inelastic interaction and phonon annihilation. These effects inhibit heat transport perpendicular to the layer interfaces [4–7]. Phonons are absorbed and dissipated along the layer interfaces. Hence, the cross plane thermal conductivity decreases. The increase of the density of electronic states due to the periodic potential barriers of nanoclusters produced by ion bombardment also increases the cross plane Seebeck coefficient and the cross plane electric conductivity. The thermoelectric figure of merit of the multilayer structure increases.

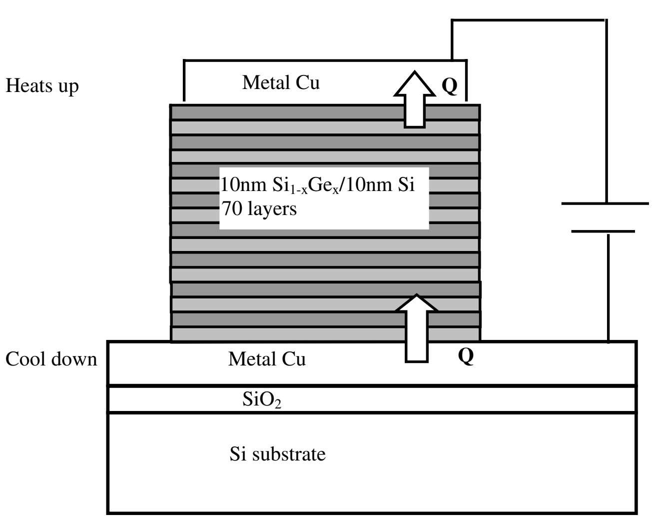

Fig. 1. Schematic of thin film TE device.

2. Experiments

2.1. Fabrication of thin film thermoelectric sample

At CIM, the electron beam evaporation deposition system with two guns was used to deposit $Si_{1-x}Ge_x$/Si multilayer thin film. The vacuum chamber with a cryogenic pump was maintained at 2×10^{-6} Torr. The multilayer films were sequentially deposited on a Si substrate, which was coated with a SiO_2 insulation layer and a metal (Cu) contact layer in advance. The thickness of the deposited layers was controlled by a crystal oscillator deposition monitor. Each Si layer was deposited at a constant rate by a single e-beam evaporator. The Si deposition rate was maintained constant while an additional evaporator periodically provided Ge atoms at a rate that determined x in alternate layers of $Si_{1-x}Ge_x$.

This technique produced a periodic structure consisting of 70 alternating layers of $Si_{1-x}Ge_x$/Si. The period was about 10 nm. Five MeV Si ion bombardment was performed on this multilayered sample using the AAMU Pelletron ion beam accelerator. SRIM simulation software [8] shows that 5 MeV Si ions pass through $Si_{1+x}Ge_x$/Si multilayer film and terminate deep in the substrate. A second Cu contact layer was later deposited over the thin film after ion bombardment. The sample was cut by a diamond cutter to get a clean edge of each layer. In this way, a complete thermoelectric sample was made.

In order to determine the stoichiometry of the $Si_{1-x}Ge_x$ layer grown in this condition, a single layer $Si_{1-x}Ge_x$ thin film was grown on glassy polymeric carbon (GPC) for the purpose of Rutherford backscattering spectroscopic (RBS) analysis. The RBS spectrum of this $Si_{1-x}Ge_x$ layer grown on GPC and the RUMP (RBS Analysis and Simulation Package) simulation results shown in Fig. 2 indicates that the $Si_{1-x}Ge_x$ may be characterized as $Si_{0.84}Ge_{0.16}$.

The Raman scattering spectrum of the 70-layer Si/$Si_{1-x}Ge_x$ thin film (10 nm each layer) before and after Si ion bombardment shown in Fig. 3 indicates that ion bombardment breaks the $Si_{1-x}Ge_x$ bonds forming nano-scale Ge and Si polycrystals and a more disordered structure.

2.2. Measurements of thermoelectric properties of thin films

2.2.1. Cross plane thermal conductivity

We used the 3ω technique to measure the cross plane thermal conductivity of the thin film as shown in Fig. 4. A narrow Pt strip, used both as heater and as a temperature sensor, is deposited on the thin film [9]. An AC current $I\sin\omega t$ heats the metal strip through two terminals by providing a known power $I^2R\sin^2\omega t$, where R is the resistance of the heater. A voltage signal IR is obtained through the other two terminals and compared in a Kelvin bridge

$$k = \alpha\frac{I^2R}{4\pi l}\left[\frac{\ln\frac{\omega_2}{\omega_1}}{\frac{V_{31}-V_{32}}{IR}}\right]\text{W/K m.} \quad (1)$$