Study of Phononic Contribution in Reduction Of Thermal Conductivity of Nano scale Superlattice Structured Thermoelectric Materials

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Abstract: ***Thermoelectrics have been receiving attention for power generation from renewable sources and waste heat because of ever increasing global demand for cost-effective and pollution-free forms of energy conversion. High performance thermoelectric materials are in search for efficient thermoelectric conversion. Superlattices have extensively generated attention as promising materials to develop thermoelectric materials with high value of figure of merit. Previous researches show low value of thermal conductivity in the direction perpendicular to the planes of Nano scale superlattice structures due to the thermal transport. Phonons in the nano scale superlattice structures are the factor which are responsible for the thermal transport that do not reside in bulk thermoelectric materials. Size of nanostructured materials, phonon mean free path and phonon wavelength are the main factors for reducing lattice thermal conductivity. A brief discussion on the phonon engineering in thermoelectric materials of superlattice structure to reduce the lattice thermal conductivity of these materials, has been presented in this paper. The research works carried out by different researchers on development of efficient thermoelectric materials having low thermal conductivity has been thoroughly reviewed in this paper.***

Keywords: Superlattices(SL), phonon blocking, nanoscale, phonon boundary scattering, thermoelectrics.

1. Introduction

Now a day a new era of nanostructured materials for thermoelectric energy conversion is approaching. One of the applications of thermoelectric materials is power generation from the waste heat which is the only means to convert thermal energy to electric energy, therefore development of highly efficient thermoelectric materials having high thermoelectric figure of merits are required. The figure of merit *ZT* for a thermoelectric material is given by

$$Z = \frac{S^2\sigma}{k},$$

Where *S* is the Seebeck coefficient, σ is the electrical conductivity, κ is the total thermal conductivity, and *T* is the absolute temperature. Thermal conductivity κ is the combination of contributions from electrons (κ_e), holes (κ_h), phonons (κ_{ph}), and bipolar diffusion (κ_{eh}) such that $\kappa = \kappa_e + \kappa_h + \kappa_{ph} + \kappa_{eh}$. Phonon-phonon scattering, phonon group velocity and phonon mean free path in nanostructured thermoelectric materials are the factors due to which κ_{ph} value decreases as compared to bulk thermoelectric materials. Many researches have been going on for attempts to reduce the phonon thermal conductivity κ_{ph} by phonon boundary scattering. Thin film superlattices are the approach of many researchers to reduce κ_{ph} inspite of no negative impact on the power factor $S^2\sigma$ which would increase *ZT* above the value of bulk materials. [1]. Phonons boundary scattering dominates over electron boundary scattering at the interface in nano structured materials therefore Nano scale thermoelectric applications are of great interest now a day. Many researches are in progress to understand the heat conduction in low dimensional thermoelectric materials [2]. Metallic oxides are good candidates for various thermoelectric applications as they have low heat conductivity, low toxicity, bearable at high temperature [3].

To formulate the problems of global warming and low power generation we require highly efficient thermoelectric materials to generate high range of power from waste heat. This paper discusses the contribution of phonons in thermal transport in nano scale superlattices.

2. Brief Survey of Literature

To improve the *ZT* values of thermoelectric materials many work has been done by doping, Bi-Doped $Mg_2Si_{0.8}Sn_{0.2}$ (Qiang Shen *et al.*, 2010 [4]); La-Doped Europium Titanate (Muta *et al.*, 2005[5]); Ca-Doped $(ZnO)_mIn_2O_3$ (Kaga *et al.*, 2004[6]); Bi-doped PbTe (Nolas *et al.*, 2011[7]); La- or Nb-doped SrTiO3 (STO) bulk single crystals (Koumoto et al., 2005[8]); $SrTi_{0.8}Nb_{0.2}O_3$ ceramic (Kato *et al.*, 2007[9]), but many researchers have observed that Nano-structuring has led to materials with the highest thermoelectric energy conversion efficiency. Hicks and Dresselhaus [10], [11] works suggested that low-dimensional materials exhibit improved thermoelectric power factor. The enhancement in phonon interfacial scattering in a Nano size materials reduce phonon thermal conductivity. Thermal transport behavior depends upon the different wavelengths of phonons [12]. The scattering of the medium and long-wavelength phonons effectively reduce the phonon thermal conductivity for thermoelectric material [13]. Medium or long wavelength phonons are scattered less effectively in alloys whereas due to boundary scattering in nanoscale materials these phonons scattered more effectively in these nanoscale materials and contribute large reduction in phonon thermal conductivity [19]. If the structure dimensions is very small as compared to phonon mean free path the dispersion branches becomes flat in size which results in decrease in phonon group velocity and increase in phonon scattering on defects and in Umklapp scattering [27]–[28]. Increase in phonon scattering and Umklapp scattering cause reduction in thin films or nanowires in-plane thermal conductivity [29]. The thermal conductivity in superlattice periodic structures is reduced due to the formation of phonon bandgaps [14]. The thermal conductivity of short period (a few nm) structures of Si/Ge superlattice [15], [16], GaAs/AlAs superlattices [17], Bi_2Te_3/Sb_2Te_3[18] can be lower than those of their respective solid solution alloys. A. Khitun et al. [26] theoretically investigated that the scattering of acoustic phonons on quantum dot superlattice effectively reduce in-plane lattice thermal conductivity of a quantum-dot superlattice. Thermal transport in semiconductors is mostly carried by acoustic phonons. Theoretical study based on first-principles calculations and semi-classical Boltzmann theories were used to investigate cross plane thermal conductivity of Bi_2Te_3/Sb_2Te_3 superlattices. Phonon thermal conduction reduces due to phonon-blocking at the interfaces of the superlattice. [30] Thus due to combine effects of decrease in phonon group velocity, increase in phonon boundary scattering, Umklapp scattering, interfacial phonon-blocking and phonon band gap formation in superlattice periodic structures, thermal conductivity decreases in these materials. Bi_2Te_3/Sb_2Te_3 and PbTe/PbSe [20]–[22], films and Si nanowires [23], [24] show reduction in lattice thermal conductivity due to phonon scattering. Nanostructured thin-films and wires do exhibit low lattice thermal conductivities which results in higher material *ZT*, Nano materials that manage scattering of phonons at all length scales are the promising materials to design for future thermoelectrics. [25]

Liu et al. [31] measured the ZT value of 0.047 of Si(20Å)/Ge(20Å) superlattice which is about 4 times higher compared to bulk Si value [32].He also observed the reduction in thermal conductivity for both in-plane and cross-plane direction. The reduction in thermal conductivity of superlattice is due to partial diffuse scattering of phonons. B. Yang et al. [33] compare the thermoelectric properties of Si 80 Å /Ge 20 Å superlattice between cross-plane and in-plane direction and observed that the thermal conductivity of these SL in cross-plane direction is 5-6 times lower than that of the in-plane thermal conductivity.The anisotropy of the thermal conductivity is mainly attributed to the interface scattering [34] and SL phonon velocity [35]-[37].

Majumdar et al. [38] studied the thermal conductivity behavior of Si/alloy superlattice and alloy/ alloy superlattice and comparison between them. He found that the thermal conductivity of $Si/Si_{0.7}Ge_{0.3}$ superlattice, decrease with the decrease in period thickness while there were no dependence on period thickness for $Si_{0.84}Ge_{0.16}$ /$Si_{0.76}Ge_{0.24}$ superlattice, which is due to alloy scattering. It was observed that interfacial acoustic impedance mismatch is responsible for the difference in the thermal conductivity behavior between these two superlattices. The AIM partially determines the fraction of phonons reflected at each interface [39]. AIM is much larger for $Si/Si_{0.7}Ge_{0.3}$ than for $Si_{0.84}Ge_{0.16}$ /$Si_{0.76}Ge_{0.24}$ .The larger AIM of superlattices increase phonon scattering. The thermal conductivity of Si/alloy superlattice is affected due to interfacial scattering however alloy/alloy superlattice is affected due to both, alloy scattering and the influence of the AIM. Alloy scattering and the influence of the AIM both are the future aspects to investigate the high AIM alloy/alloy superlattice which shows combine effect on the thermal behavior of alloy/alloy superlattice.

A. Majumdar et al. [40] investigated the thermal conductivity of Si/SiGe superlattice nanowire, he suggested that thermal conductivity can be reduce by the contribution of phonons in boundary scattering, In these superlattice nanowires alloy scattering of phonons also contributes thermal conductivity reduction. Nanowire boundary scattering causes scattering of long-wavelength acoustic phonons and alloy scattering of short wavelength acoustic phonons in SiGe alloy is due to imperfections in the alloy segments. Scattering of both type of phonons having short and long wavelength control the heat transport in Si/SiGe superlattice nanowire. He observed that the behaviour of Si/SiGe nanowire in reduction of thermal conductivity is similar to 2D superlattice thin film behavior in which thermal conductivity increases upto 200 K. However, thermal conductivity of nanowire is less than that of the $Si/Si_{0.7}Ge_{0.3}$ superlattice thin film.

In the previous work on Bi_2Te_3 /Sb_2Te_3 superlattice , Goodson et al. [41] found that the thermal conductivity to be significantly lower than the bulk value for Bi_2Te_3. The reduction of thermal conductivity for small period superlattices is due to scattering at interfaces and intrinsic phonon-phonon scattering. Phonons responsible to reduction of thermal conductivity of the Bi_2Te_3 /Sb_2Te_3 superlattice layers have low frequency with longer mean free paths.

Z. Xiao et al. [42] observed that the nanostructured Bi_xTe_3/Sb_2Te_3 thin film multilayer having alternate periodic structure of eleven or thirty-nine with the thickness of each layer 10 nm has low thermal conductivity as compared to their bulk materials. He calculated the value of thermal conductivity of 6.2mW/cm K for the multilayer film with 11 layers and of 5.1mW/cm K for 39 layers and found that thermal conductivity decreases with increasing layers. Values of ZT as high as 2.4 have been measured in p-doped Bi_2Te_3/Sb_2Te_3 superlattices[43] However, Bi_2Te_3/Sb_2Te_3 superlattices contain poisonous elements like bismuth or lead and they decompose at T≈200 C where main working of thermoelectric materials are expected, these materials are not suitable for environment friendly thermoelectric applications[44].

Fernandez et al. [44] theoretically found a large Seebeck coefficient in 2DEG confined in $(SrTiO3)_5$/ $(SrRuO3)_1$ superlattice, S =1500μV/K, larger than that found in bulk Nb-doped SrTiO3 thin films. Ohta et al. [45] found that a very high value of S= 850 μV/K and ZT of 2.4 for the 2DEG was estimated for periodic two dimensional electron gas (2DEG) $SrTiO_3$/Nb-doped $SrTiO_3$ superlattices formed at the Nb-doped layer, when thickness decrease to one unit cell.

In the recent work [46] on n-type $LaAlO_3/SrTiO_3$, due to electronic confinement at the interface, there is no enhancement of Seebeck effect is observed. Previous researches in the search of highly efficient thermoelectric materials at high temperatures have been growing interest towards metal oxides such as $Na_{0.75}CoO_2$ (p type, $ZT_{300\ K}$~0.1) [47], $SrTiO_3$ (n-type, $ZT_{300\ K}$~0.08) [48] and $Ca_3Co_4O_9$ (p-type, $ZT_{300\ K}$~0.07) [49]

Oxide SLs. are considered as efficient thermoelectric materials therefore Choi et al. observed Nb:STO/STO SLs, in which he concluded that Seebeck coefficient of Nb:STO/STO SLs increases with decreasing Nb:STO layer thickness and reaches to almost 500 μV/K when thickness decrease to 1 unit cell[51] The polaron which is a quasi-particle is a coupling of electron and phonons which are responsible to enhance the seebeck coeeficient in oxide superlattice due to increased effective mass of these particles in these oxide superlattice.[52]

H.Ohta et al. [50] found that two-dimensional electron gas (2DEG) confined within $(SrTiO3)_{16}/(SrTi_{0.8}Nb_{0.2}O_3)_1$ superlattices exhibit Seebeck coefficient of 320 μVK^{-1} at room temperature which is about 5 times higher as compared to bulk $SrTi_{0.8}Nb_{0.2}O_3$ of 60 μVK^{-1} and hence the higher ZT value of this superlattice. This work suggests that 2DEG SrTiO3-based thermoelectric materials are the attractive thermoelectric materials for the future aspects.

Y. Chen et al. [52] investigated the effects of interfacial scattering, nanowire boundary scattering, and period length in Kr/Ar nanowires. Thermal conductivity can be reducing by the phononic contribution in boundary scattering [40]. The lattice thermal conductivity that is only one-third of that of pure Ar nanowires is due to the interfacial thermal resistance in the Kr/Ar nanowires .The thermal conductivity of 2D Si/SiGe superlattice films was more than the thermal conductivity of Si/SiGe superlattice nanowires [53], [54]

W. E. Bies et al. [55] calculated the three-fold thermal conductivity reduction due to phonon dispersion in HgTe/CdTe SLs whose data shows a reduction factor in κ/τ of 3.0 , where κ is the lattice thermal conductivity and lifetime . This factor reflects phonon-dispersion effects only.

M. Zebarjadi et al. [56] studied the Metal/semiconductor superlattices as these superlattices are efficient thermoelectric materials of high *ZT* value. He investigate theoretically and experimentally, the thermoelectric transport properties of superlattices structure of ZrN/ScN metal/semiconductor .Boltzmann transport model was developed to theoretically calculate the thermoelectric properties of a superlattice structure.[57] Figure of merit ZT of 1.5 at 1300 K was found theoretically for ZrN/ScN superlattice structure. Interface scattering of phonons can decrease thermal conductivity more effectively than that of electrons as phonon blocking in superlattices [58]. The cross-plane lattice conductivity of 1.8 W/m K measured at room temperature was used to calculate the thermoelectric properties of ZrN/ScN superlattice [59]. With the increase in temperature, reduction of lattice conductivity was expected. There are many thermoelectric materials which are very efficient at low temperature below 500ºC, but do not shows high figure of merit at high temperatures due to their instabilities. A metal/semiconductor structure was designed and grown for high-temperature applications high figure of merit at high temperatures.

T. Borca-Tasciuc et al. [60] observed that the thermal conductivity of InAs/AlSb peaks around 150 K. The thermal conductivity reduces after this peak temperature which shows the interfacial effects on thermal conductivity in SLs structure at higher temperatures. The values of thermal conductivity of InAs/AlSb superlattices measured are lower than those of the corresponding bulk structure and decrease with the increase in temperature.

J. Piprek et al. [61] investigations of GaAs–AlAs superlattices indicate strong interface scattering of phonons at the interface which exhibit a strong reduction in thermal conductivity parallel to the interface of 0.35 ±0.05 W/cm K.

Yang Yu-Rong et al. [62] studied the thermal conductivity of GaAs/AlAs theoretically and observed that it increases with the increase of temperature below 60K at which it showed a peak behavior and reduce at high temperatures which explain that due to zone folding in superlattices, the phonon group velocity decrease which further cause the reduction of heat conduction in these superlattice structures [63, 64]. Phonons are responsible for the conduction of heat in superlattices, therefore the formula for the lattice thermal conductivity κ at any temperature T is given as [64], [65]

$$\kappa(T)=\Sigma_{\lambda}\tau_{\lambda}(T)C_{\lambda}(T)V_{g\lambda}^{2},$$

where τ_{λ}, C_{λ}, and $V_{g\lambda}$ are the phonon relaxation time, specific heat and phonon group velocity having phonon state λ, respectively.

The observable mechanism in the conduction of heat in superlattices is phonon boundary scattering at relatively low temperature and Umklapp scattering process which dominates when the temperature increases. The calculated value shows that both of these mechanisms increase the heat conduction in superlattices.

3. Conclusions

The research efforts made by different researchers to understand the phononic engineering in thermal transport in different superlattice structured thermoelectric materials, have thoroughly reviewed in this paper.

The unexpected reduction of the thermal conductivity is due to low frequency acoustic phonons with longer mean free paths. We expect that the scattering of heat-carrying phonons on well/barrier boundaries can reduce the thermal conductivity at superlattice periods of several nanometers. From this, we conclude that the thermoelectric figure of merit can be increase by reducing thermal conductivity in superlattice structures. Thermal conductivity of the superlattices layers was found to be significantly lower than the bulk value.

Thermoelectric with low thermal conductivity or high figure of merit have been receiving much attention for power generation from renewable sources and waste heat because of ever increasing global demand for cost-effective, pollution-free and eco-friendly forms of energy conversion.

Among all the thermoelectric superlattices, oxide based superlatices have been growing much promising materials for future aspects in thermoelectric as these materials have low thermal conductivity, non-toxic and bearable at high temperature. We hope that this review will help us to understand the phononic contribution in the superlattice to reduce thermal conductivity and hence high figure of merit.

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