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TO STUDY THE THERMOELECTRIC PROPERTIES OF LOW DIMENSIONAL STRUCTURES

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Abstract: - In this paper we have shown that a finite acoustic mismatch between structure and barrier materials in low-dimensional structures leads to the acoustic phonon confinement, which in its turn brings about a corresponding decrease of the phonon groups velocity and modification of the phonon density of states. These factors contribute to the reduction of the in-plane lattice thermal conductivity, thus allowing one to increase the thermoelectric figure of merit. Results of experimental study of confined acoustic phonons in single Si thin films and Si/Ge superlattices are also reported High-resolution Raman spectroscopy of ultra-thin silicon-on-insulator structures reveals multiple peaks in the spectral range from 50 cm^{-1} to 160 cm^{-1}. The peak position is consistent with the theoretical predictions and indicate the confined nature of phonon transport in thin films and superlattices with a finite acoustic mismatch between layers. This opens up a novel tuning capability for optimization of the thermoelectric properties of low-dimensional structures.

Keywords: Wiedmann-Franz law, Superlattices, Raman spectroscopy

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INTRODUCTION

It is customary to express the usefulness of a thermoelectric material for refrigeration or power generation applications in terms of the dimensionless quantity ZT where T is the temperature and Z is the thermoelectric figure of merit

$$Z = \frac{S^2\sigma}{K} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Here S is the thermoelectric power, σ is the electrical conductivity and K is the thermal conductivity. Large values of ZT require high S, high σ, and low K. Since an increase in S normally implies a decrease in σ because of carrier density considerations, and since an increase in σ implies an increase in the electronic contribution to K as given by the Wiedemann-Franz law, it is very difficult to increase Z in typical thermoelectric materials [1].

It has been shown theoretically that confinement of acoustic phonons in semiconductor structures brings about significant modification of their thermoelectric properties [2-4]. The predicted increase of the thermoelectric figure of merit in quantum wells and superlattices with distinct boundaries has been a result of a significant drop of the phonon group velocity due to spatial confinement [4]. The latter leads to an increase of the phonon relaxation rate and thus, to a strong drop of the in-plane lattice thermal conductivity. This modification of the thermoelectric figure of merit ZT comes in addition to earlier predicted increase due to two-dimensional confinement of carriers [5], and increased phonon-boundary scattering in thin films [6] and in quantum wires [7]. The perpendicular thermal transport in Si/Ge super lattices was also shown to be suppressed at high temperature due to the acoustic mismatch at the boundaries [8].

The boundaries of low dimensional structures are either free or clamped-surface boundaries. The former corresponds to a free standing thin film, the latter corresponds to the thin film embedded within rigid material. Most of the real experimental situations fall into the category of intermediate boundary conditions, which allow for partial phonon wave function penetration through the boundaries. Quantitatively the difference in the 'rigidity' of materials can be characterized by the acoustic mismatch $K=\rho_2V_2/\rho_iV_i$, where ρ_i is the density of ith material and V_i is its sound velocity. Thus, it is important to establish whether acoustic phonons are indeed confined in a thin semiconductor film embedded within material of finite acoustic mismatch.

Previously, modification of acoustic phonon modes has been extensively studied in superlattices. Such modification was evident by appearance of the folded phonon doublets in Raman spectra [10]. These doublets originate due to additional periodicity of the superlattices, and can be theoretically described by Rytov's model [11]. To the best of our knowledge, spatial confinement of acoustic phonons in Raman spectra of a single thin film has not been demonstrated yet. This is primary due to the lack of high quality thin films with sharp interfaces embedded within material with distinctively different elastic properties. In most of cases, one deals with quantum wells growth and material with very similar crystalline structure.

The effects of phonon confinement on the thermal properties of low-dimensional structures also still await experimental investigation. In this paper we report data indicating confined nature of propagating phonon modes in a single Si thin films. We also establish correlation between the strength of phonon folding in Si/Ge superlattices and their thermoelectric properties.

1. Sample Preparation and Measurements

In order to prove confined nature of acoustic phonons in semiconductor thin films, we have studied ultra-thin silicon-on-insulator structures specially prepared by wafer-bonding technique. The -state -of -the- art technology allowed us to fabricate ultra thin Si films with thickness W=30nm,-90nm, and very sharp boundaries. The film were embedded within materials of significantly different elastic and crystalline properties such as Sio_2. Thermal conductivity of Sio_2 varies from 0.66 W/mK to 1.4 W/mK, as compared to 148 W/mK for bulk Si. The ultra-thin silicon-on-insulator structures are ideal for study of confined phonon thermal transport since the heat flux mostly propagate in the in-plane direction, and the acoustic phonon modes are confined due to nano scale width.

We have also examined small period Si/Ge superlattice structures grown on a p-type Si(100) wafer. A typical structure consists of a buffer layer, and 150 periods of 33ÅSi/33ÅGe superlattice with a uniform heavy n-type doping.

2. Raman Spectra of Ultra-Thin Silicon-on-Insulator Structures

A typical spectrum of silicon-on-insulator structure with 30nm thick Si layer is shown in Figure 1. In addition to easily recognizable Si peaks at 522 cm^{-1} (TO), 970 cm^{-1}(2TO), 434cm^{-1}(LO), and 302 cm^{-1} (2TA/LA), we have also observed quasi-equidistant peaks in the low-frequency end of the spectrum, in the range from 50cm^{-1} to 160cm^{-1}. The peaks below 50cm^{-1} have been cut by the Raman spectrometer filter.

In order to exclude local vibrational modes of SiO_2 from consideration, we have carried out Raman spectroscopy of Si substrates with the layers of SiO_2. In this case, no peaks were observed in the specified frequency range from 50cm^{-1} to 160cm^{-1}. The investigation was performed for different samples to make sure that the presence or absence of peaks is not related to the finite penetration depth of the incident laser light.

The spectral position of the additional quasi equidistant peaks depended on the thickness of the Si layer embedded within layers of SiO_2. Figure 2 presents a blow-up of these peaks with the exact values of the peak position. In Table 1 we present experimental values of phonon peaks extracted from the Raman spectrum of one of the samples, and a theoretical fit based on a calculated phonon dispersion for a Si thin film of given thickness [3], Due to spatial confinement effects bulk acoustic phonon branches (LA and TA) split into many confined phonon modes [3-4]. The calculated dispersion relations for thin Si film are shown in Figure 3. Raman spectrometer probes these modes for the phonon wave vector q close to the center of the first Brillouin zone center.

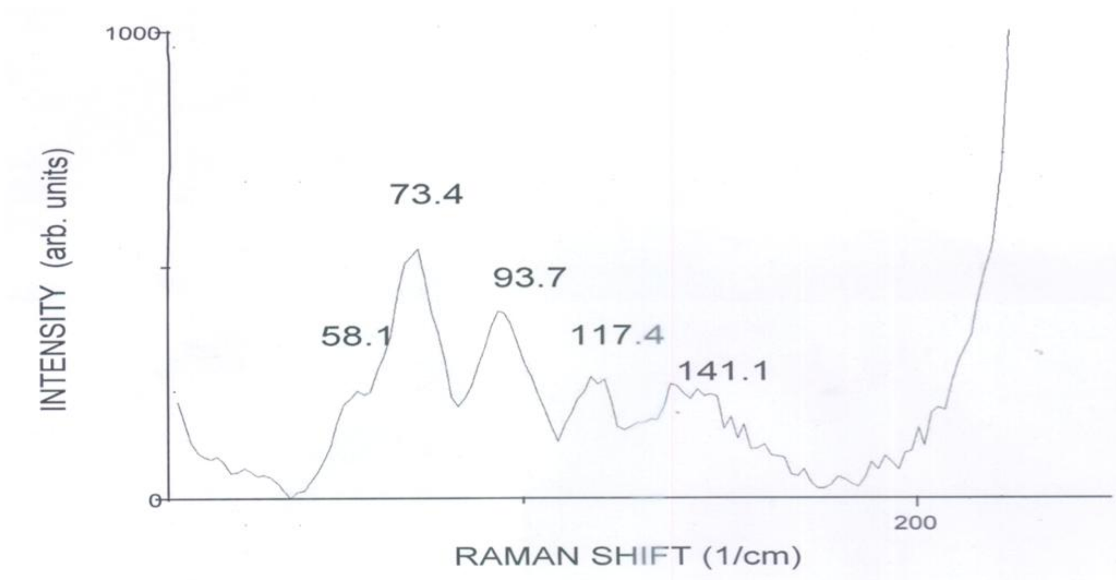

Figure- 1. Low frequency tail of the Raman spectrum of the ultra-thin BESOI. The peaks below 50 cm^{-1} are cut by the Raman spectrometer filter. The peak position changes with the thickness of the si layer.

As one can see, the calculated values of the peak positions for a 30nm wide thin film are in good agreement with the measured ones.

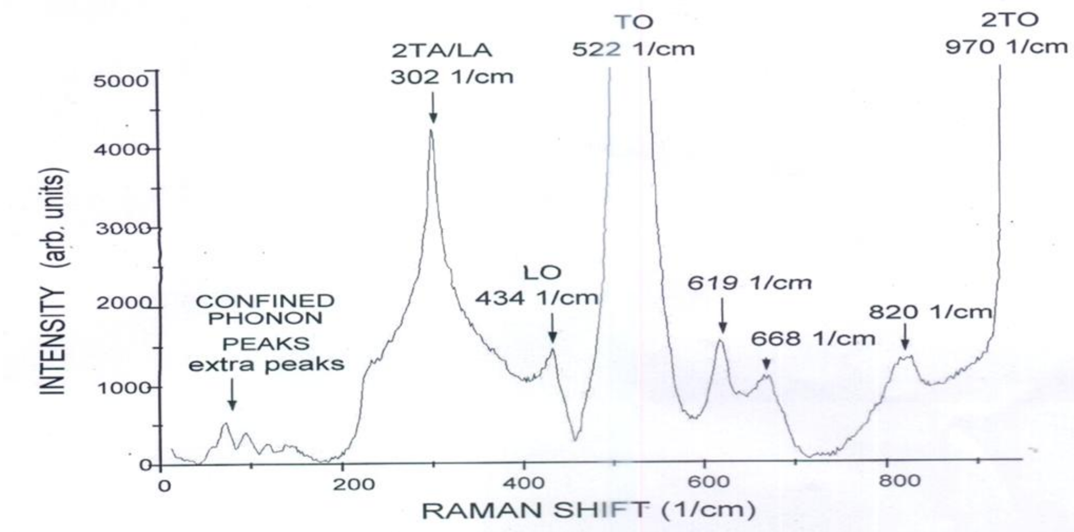

FIGURE- 2. Raman spectrum of the ultra-thin BESOI structure. The thickness of the silicon layer is W=30nm. Additional peaks in the range from 50cm^{-1} to 140 cm^{-1} have been attributed to the confined acoustic phonons.

Thus, we may conclude that the observed additional peaks in the low-frequency end of Raman spectra from ultra-thin silicon-on-insulator structures are indeed related to spatially confined acoustic phonon modes. These confined phonon peaks originate from a single thin film rather than from a superlattice. They are described by different dispersion relation [3] and, in this sense, they have different origin from that of folded doublets in Raman spectra of superlattice structures [10] which are described by Rytov's model [11].

TABLE- 1 Experimental and Theoretical Peak Position

E^{exp}, meV	1.2	1.5	1.9	2.4	2.8
E^{mod}, meV	1.1	1.5	1.9	2.2	2.6

The fact that acoustic phonons are at least partially confined in low-dimensional structures with finite acoustic misfit indicate that the phonon quantization have to be included in the modeling of phonon transport in such structures. The important consequence from this in that the effective phonon group velocity will be lower in the low dimensional structure than in the corresponding bulk material [3-4]. The amount of the velocity decrease, and corresponding increase in phonon relaxation rate, can be "phonon engineered" by appropriate change of the film thickness and acoustic mismatch K of the boundary material.

4. Thermal Conductivity of Si/Ge Superlattices

Phonon quantization observed in Raman spectra of ultra-thin silicon-on-insulator structures with sharp boundaries should have strong effect on thermal transport in these structures. The measurement of the in-plane thermal conductivity of silicon-on-insulator structures is currently in progress.

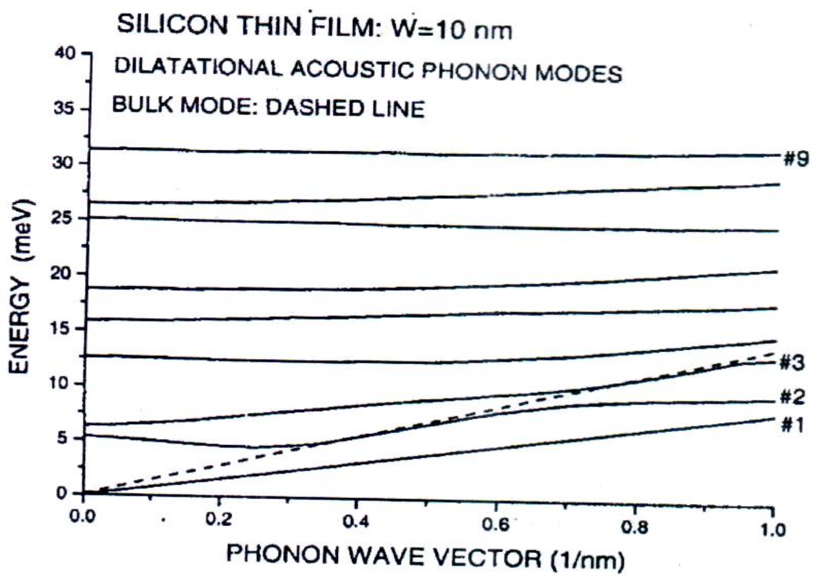

FIGURE- 3. Confined dilatational acoustic phonon modes in a 10 nm wide thin Si film. For comparison, the dashed line shows the bulk dispersion. Note that the slopes of the phonon dispersion branches, which define the group velocity, are smaller in the thin film than in the bulk.

This work is significantly complicated by the very small thickness of the Si layers. The smallest silicon-on-insulator structure, for which the in-plane thermal conductivity data was reported [13], had a thickness on the order of 100 nm. It was found that phonon-interface scattering reduces the thermal conductivity by up to 50% at room temperature.

As an intermediate step, we have measured thermal conductivity of Si/Ge superlattices. This allowed us to examine a correlation between modification of phonon modes in low dimensional structures and their thermal properties. The acoustic misfit between Si and Ge is about K=0.75. Thus, we can expect that changing thickness of Si and Ge layers and mole fraction in mixed $Si_x Ge_{1-x}$ layers, one can re-engineer phonon propagation characteristics in a rather wide range.

Details of Raman spectroscopic investigation of SiGe superlattices with different period D have been reported earlier [12]. Knowing the folded peak frequency ω_m and folding index m, we can determine the effective phonon velocity from Rytov's model $V_{eff} = \omega_m D/(2\pi m)$ [11]. The smallest velocity $V_{eff} = 1.5\times10^5$ cm/sec , which is much less than expected from the bulk values, has been obtained for Si/Ge superlattice with 150 periods of 33Å Si/33ÅGe layers.

Thermal conductivity measurements have been carried out using the 2 wire-3ω method. The measured thermal conductivity of the superlattice of the sample with D=3 nm was 1.8W/mK in the cross-plane direction. The cross-plane thermal conductivity of the buffer layer was determined to be 7.5 W/mK, obtained thermal conductivities were considerably lower than those determined using the bulk thermal conductivities for Si, Ge, and $Si_x Ge_{1-x}$ alloys [14]. The strongest drop in thermal conductivity corresponded to the lowest phonon group velocity, and could be attributed to the modification of the phonon modes, which manifests itself as phonon folding in the Raman spectra. Currently we are measuring the in-plane thermal conductivity of Si/Ge superlattices using 2 wire-3ω method.

Confinement of phonon modes strongly increases phonon relaxation but does not significantly increase phonon-electron scattering rates [3-4]. Due to this reason, one can realize "electron transmitting- phonon blocking" transport regime, which leads to ZT increase. The results of our investigation show that by changing thickness of semiconductor layers and their acoustic mismatch with the boundaries, we can optimize the thermoelectric properties of low dimensional structures via phonon engineering.

5. CONCLUSIONS

We have demonstrated confined nature of acoustic phonon modes in a single semiconductor quantum well by carrying on high resolution Raman spectroscopy of ultra-thin silicon-on-insulator structures. The obtained data indicate multiple confined acoustic phonon peaks in Raman spectra of the silicon-on-insulator structure with the thin film thickness of 30nm. It was also shown that position of the folded doublets from the longitudinal acoustic phonons in Si/Ge superlattices indicates the strength of phonon confinement in these structures and correlates with their in-plane lattice thermal conductivity. The experimental results are consistent with the recent theoretical predictions of strong decrease of the lateral lattice thermal conductivity due to phonon confinement. Engineering of phonon modes via selective spatial confinement opens up an additional tuning capability for optimizing of the thermoelectric properties of semiconductor structures.

REFERENCES

1. H.J. Goldsmid, Electronic Refrigeration (Pion, London, 1986).

2. A. Balandin and K.L. Wang, Nanostructures : physics and Technology, edited by Zh. Alferov, L. Esaki (Ioffe Institute, St. Petersburg, Russia, 24 (1998)

3. A. Balandin and K.L. Wang, Phys. Rev. B 58, 1544 (1998).

4. A. Balandin and K.L. Wang, J. Appl. Phys., 84, 6149 (1998).

5. L.D. Hicks and M.S. Dresselhaus, Phys. Rev. B 47, 12727 (1993).

6. G. Chen, Phys. Rev. B 57, 14958 (1998).

7. S.G. Walkauskas, D.A. Broido, K.Kempa and T.L. Reinecke, J. Appl. Phys., 85, 2579(1999).

8. P. Hyldgaard and G.D. Mahan, Phys. Rev. B, 56, 10754 (1997).

9. G.D. Mahan and M. Bartkowiak, Appl, Phys., Lett., 74, 953(1999).

10. C. Colvard, T.A. Gant, M.V. Klein, R. Merlin, R. Fischer, H. Markoe, and A.C. Gossard, Phys., Rev. B 31, 2080 (1985).

11. S.M. Rytov, Soviet Phys. JETP, 2, 466 (1956).

12. J. Liu, A Balandin, T. Borca-Tasciuc, Y.S. Tang, K.L. Wang and G. Chen, Proc. of the Materials Research Society, Boston, USA, November 1998.

13. Y.S. Ju and K.E. Goodson, Appl. Phys Lett., 74, 3005 (1999).

14. T. Borca- Tasciuc, D. Song, J.L Liu, G. Chen, K.L. Wang, X. Sun, M.S. Dresselhaus, T. Radetic, and R. Gronsky, Proc. of the Materials Research Society, Boston, USA, November 1998.