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Acoustic Phonon Engineering of Cubic Semiconductor Quantum Dot

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Abstract

Phonon spectrum in nanostructure such as quantum well, quantum wire and quantum dot can be modified due to quantum confinement effect. Elastic continuum approach with finite difference method can be used to calculate the phonon dispersion of semiconductor cubic quantum dot from which Phonon group velocity can also be found by numerical differentiation. In the present work, we will theoretically investigate the effect of coating made of acoustically different material on phonon spectra and phonon group velocity. Coating materials may be acoustically fast or acoustically slow than the quantum dot's material and we will observe how they affect the phonon spectra and phonon group velocity of the quantum dot differently. Emphasis gives on the observation of changes experienced by phonon spectra in quantum dot and modification of them by barrier material leads to the concept of phonon engineering.

Keywords*: nanaophononics, phononstopband, acoustic phonon, quantum dot.*

1. Introduction

Today's technology of semiconductor devices has been decreased to the nanometer scale. So the physicist and engineers are stimulated for the research of understanding the novel properties of semiconductor nanostructures such as quantum well, quantum wires and quantum dot. Although there has much work on the electronic properties of nanostructures, research in phonon properties has attracted significant attention in recent years [1-6]. In a semiconductor, acoustic phonons are the dominant heat carriers. In analogy to electronic band structure, phonon dispersion is an important property that directly affects the lattice thermal conductivity of the semiconductor. One of the main factors that drive the current interest in phonon modification in semiconductor nanostructures is the increased heat dissipation associated with an increase in the transistor packing density [7]. In order to dissipate the increasing amount of heat from the chip area, one has to engineer material parameters or structure geometry in such a way that thermal conductivity is large along particular directions. To improve performance of thermoelectric, one needs to achieve low thermal conductivity. But these are two demands are contradictory and both can be approached with proper modification of phonon spectra [5]. In the present work we will investigate the acoustic phonon modification of cubic semiconductor quantum dot. A nomenclature of analytical results for phonon modes of free standing slabs can be found, for example, in [8,9]. Changes in phonon modes was first studied in 1950s. Later in 1980s and 1990s folded phonons and confined acoustic phonon modes was observed in quantum well superlattices (QWS) [10] and freestanding thin films and nanowires respectively. Most papers on the subject used the elastic continuum approach for calculating phonon dispersion and adopted solution techniques developed in acoustics and mechanics. Balandin and Wang [5] have pointed out that the

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confinement-induced changes in the acoustic phonon dispersion can lead to a much stronger effect on thermal conductivity [11]. It was later shown theoretically [6,12,13] that in the thin films (quantum wells) or nanowires (quantum wires) embedded in the acoustically different materials, the phonon group velocity as well as thermal conductivity can be enhanced or reduced along certain directions. QDS have been proposed for the solar cell, thermoelectric, photodetector, and photovoltaic applications [14-17]. The phonon modification can be used in the design of the nanoscale transistors, vertical metal-oxide-semiconductor, field-effect transistor (MOSFET), alternative-gate dielectric transistors, etc [1]. Actually in the past decade, the dynamics of acoustic phonons in QD superlattices or QD crystal has attracted a lot of attention [18-23]. Phonon spectrum has also been studied in case of graphene and other nanostructure [2, 4]. In our earlier work [24], elastic continuum approach with finite difference method was used to calculate the phonon dispersion of semiconductor cubic quantum dot from which phonon group velocity was found by numerical differentiation. In this research we will coat a cubic quantum dot with acoustically different material and observe the effect of coating on phonon spectra.

2. Theory

In case of nanostructures, we et al. [24] have used elastic continuum approach to get phonon dispersion of cubic quantum dot with cubic crystal. The equation of motion for elastic vibrations is given by

$$\rho \frac{\partial^2 U}{\partial t^2} = \frac{\partial \sigma_{ij}}{\partial x_i} \quad (1)$$

Where U = (u, v, w) is the displacement vector, ρ is the mass density of material, σ_{ij} is the elastic stress tensor. For cubic crystal the number of elastic stiffness constant reduces from 36 to 3 (C_{11}, C_{12} *and* C_{44}) considering cubic structure. The displacement vector has three components (*u, v and w*). Considering the particle displacement in x direction the equation of motion

$$\rho \frac{\partial^2 u}{\partial t^2} = C_{11} \frac{\partial^2 u}{\partial x^2} + C_{44}\left(\frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2}\right) + (C_{12} + C_{44})\left(\frac{\partial^2 v}{\partial x\, \partial y} + \frac{\partial^2 w}{\partial x\, \partial z}\right) \quad (2)$$

Similar expressions are for y and z directions. In case of nanostructure we solved the equation numerically instead of analytically with the finite edge boundary condition [6,12,13,24]. We considered a shear wave travelling in the x direction with a particle displacement in the y direction. We know that shear mode has only one nonzero component (o, v, o). Then the equation of motion would be

$$\rho \frac{\partial^2 v}{\partial t^2} = C_{11} \frac{\partial^2 \boldsymbol{v}}{\partial y^2} + C_{44}\left(\frac{\partial^2 \boldsymbol{v}}{\partial x^2} + \frac{\partial^2 \boldsymbol{v}}{\partial z^2}\right) \quad (3)$$

We look for a solution of this equation as

$$v = v_z \exp[i(\omega t - kx)]$$

Substituting the solution into equation (3) the partial differential equation will turn into an ordinary differential equation.

$$\frac{d^2 v_z}{dy^2} + \frac{1}{C_{11}} \frac{dC_{44}}{dz} \frac{dv_z}{dz} + \frac{1}{C_{11}} (\rho\omega^2 - k^2 C_{44})\, v_z = 0 \quad (4)$$

The boundary condition requires that the stress and strain are zero at the surface that means $\frac{dC_{44}}{dz} = 0$ in equation (4). To satisfy this condition we require that the boundary condition would be

$$\left.\frac{dv_z}{dz}\right|_{z=at\ the\ edge\ of\ QD} = 0$$

This boundary condition is referred to as Neumann boundary condition. Now we will apply the numerical technique to solve the differential equation (4) with the above mentioned finite edge boundary condition. To solve the equation numerically for phonon dispersion we use finite difference method and follow a general strategy for vibration analysis [6,7,12,13,24].

The numerical algorithm takes four steps. First we divide the total thickness of the QD into N equal parts of width h=a/N. Where a = the edge of quantum dot. For example in case of quantum dot of a= 10nm edge we can divide the total thickness in z direction i.e. in (001) crystallographic direction. In that case if we consider N=10, then h = 10/10=1 nm. Then the step size would be dx= dy= dz =1 nm. We define the stiffness constants and density for each node in the z direction. Now we apply the central difference formula to derivatives [3] and get

$$\frac{d^2v}{\mathrm{d}y^2} \approx \frac{v_{i+1} - 2v_i + v_{i-1}}{h^2}$$
$$\frac{dv}{\mathrm{d}z} \approx \frac{v_{i+1} - v_{i-1}}{2h}$$
$$\frac{dC_{44}}{\mathrm{d}z} \approx \frac{C_{44,i+1} - C_{44,i-1}}{2h}$$

Applying these in equation (4), we get

$$\left(\frac{-\mathrm{C}_{44,\mathrm{i}+1}+4\mathrm{C}_{11,\mathrm{i}}+\mathrm{C}_{44,\mathrm{i}-1}}{4\mathrm{h}^2}\right)\mathrm{v}_{\mathrm{i}-1} + \left(\rho_{\mathrm{i}}\omega^2 - \frac{2\mathrm{C}_{11,\mathrm{i}}}{\mathrm{h}^2} - \mathrm{C}_{44,\mathrm{i}}\mathrm{k}^2\right)\mathrm{v}_{\mathrm{i}} + \left(\frac{\mathrm{C}_{44,\mathrm{i}+1}+4\mathrm{C}_{11,\mathrm{i}}-\mathrm{C}_{44,\mathrm{i}-1}}{4\mathrm{h}^2}\right) = 0 \qquad (5)$$

The above equation represents N+1 algebraic equations with i= 0, 1, 2, 3, …….. N. Now we will apply the finite difference method to the Neumann boundary condition on equation (5). Using central difference formula we get the boundary condition

$\frac{dv}{\mathrm{d}z} \approx \frac{v_1 - v_{-1}}{2h} = 0$ and $\frac{dv}{\mathrm{d}z} \approx \frac{v_{N+1} - v_{N-1}}{2h} = 0$

Which gives $v_1 = v_{-1}$ and $v_{N+1} = v_{N-1}$ respectively. We will apply boundary conditions into these N+1 algebraic equations. The problem is now represented by a system of N+ 1 linear and homogenous equation with N+ 1 unknown ($v_0, v_1, v_2, \ldots\ldots\ldots\ldots\ldots v_N$). We can represent these equations in matrix form. We can write these in the form of $[[\mathrm{A}] - \omega^2[\mathrm{I}]\{X\} = 0$, where, $\omega^2 = \lambda$(eigenvalue). For the given parameters (C_{11}, C_{44} and ρ) we can solve the eigenvalues for each value of the wave vector q. The phonon dispersion is then plotted as a function of ω versus q. We can multiply the frequency,ω with $\hbar$ to get the dispersion relation E versus q [6, 24]. Whole procedure is implemented using MATLAB. Considering a shear wave travelling in the x direction with the particle displacement in y direction, we et al. [24] applied the finite difference method for the solution of the elasticity equation with piecewise uniform parameters in z direction of the constituent material. That means C_{11}, C_{44} and ρ are the piecewise function of z. In case of uncoated quantum dot the material parameters was the function of the constituent material for each nodes (10 nodes for 10 equal parts of quantum dot). But in coated quantum dot for the 1st, 2nd, 9th and 10th nodes would take the parameters of coating material. The remaining nodes would take the material parameters of QD.

3. Numerical Results and Discussion

Using the model we calculated the dispersion relation of individual quantum dot with edge ranging from 5 nm to 10 nm [25]. Here we have considered the edge of smallest GaAs quantum dot as 5 nm because of the breakdown of elastic continuum approach below 4 nm.

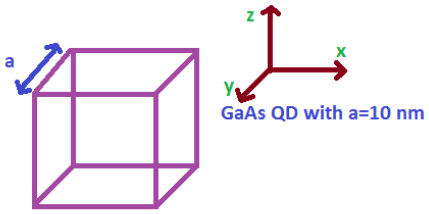

Figure 1. Schematic of a Cubic Quantum Dot

Elastic stiffness constants, $C_{11} = 12.21 \times 10^{10} \mathrm{Nm}^{-2}$ and $C_{44} = 5.99 \times 10^{10} \mathrm{Nm}^{-2}$, Density, $\rho = 5360\ \mathrm{Kgm}^{-3}$ [12,13,22]. Using these parameters we calculated the frequency ω from the solution of the eigenvalue problem of theory section [24]. Phonon dispersion for an individual cubic GaAs quantum dot with 10 nm edge, 6nm edge and 5nm edge are shown in figure 2.

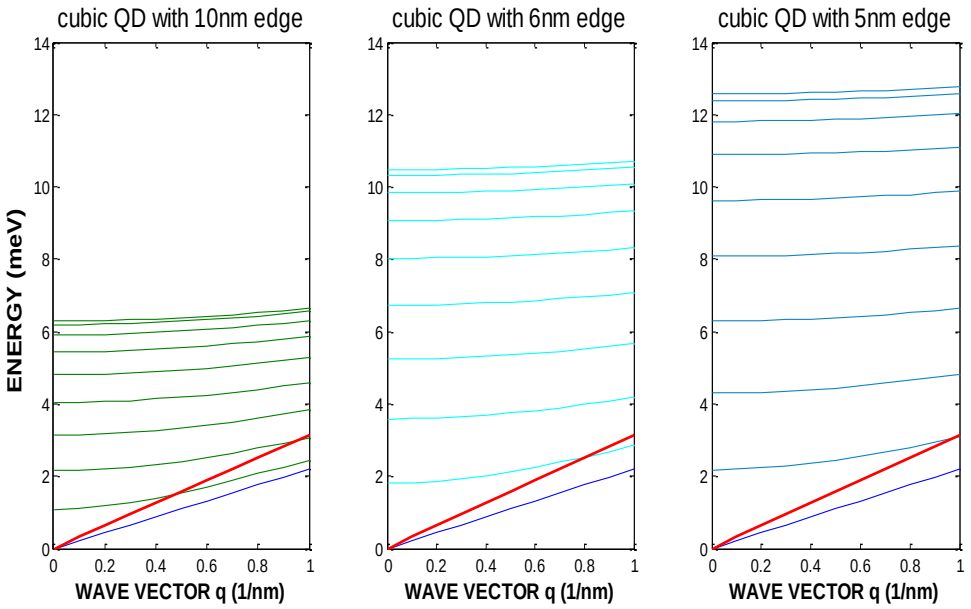

Figure 2. Phonon Dispersion Relation for Cubic GaAs Quantum Dot with 10 nm edge, 6nm and 5nm edge along with the Dispersion Relation of Bulk Phonon (shown in red line) [24]

Discrete phonon branches exhibits energy quantization similar to energy quantization of electron in quantum dot. From this figure we can see that phonon dispersion in quantum dot nanostructure changes due to phonon spatial confinement induced by the finite boundaries of the quantum dot. In quantum dot nanostructure there are two types of phonon branches. For the first branch it has a linear dispersion relation. First branch of the three different QDs is exactly similar named as surface phonon mode. For the second branch and above, there exist a cut off frequency at the center of the Brillouin zone, i.e. $\omega \neq 0$ for q = 0 and for this reason sometimes these modes are called quasioptical modes.

These quasioptical modes are different from pure optical modes and created by periodic scattering of quantum dot interfaces. Comparing the dispersion relation of the three quantum dot of different sizes we observed that energy increases as size decreases. We found the exact values of phonon group velocity for each phonon branch by numerical differentiation. Corresponding group velocities of three different quantum dots of different size are shown in figure3. From this figure we noticed that the higher the mode number the smaller the group velocity. Group velocity of the first branch is near the bulk velocity, it then drops dramatically with the increasing energy. Thus the overall group velocity decreases. The group velocity is an important property for determining other phonon properties such as phonon relaxation rate, phonon density of states, phonon scattering etc. Group velocity directly affects the lattice thermal conductivity, so modification of group velocity will modify the lattice thermal conductivity.

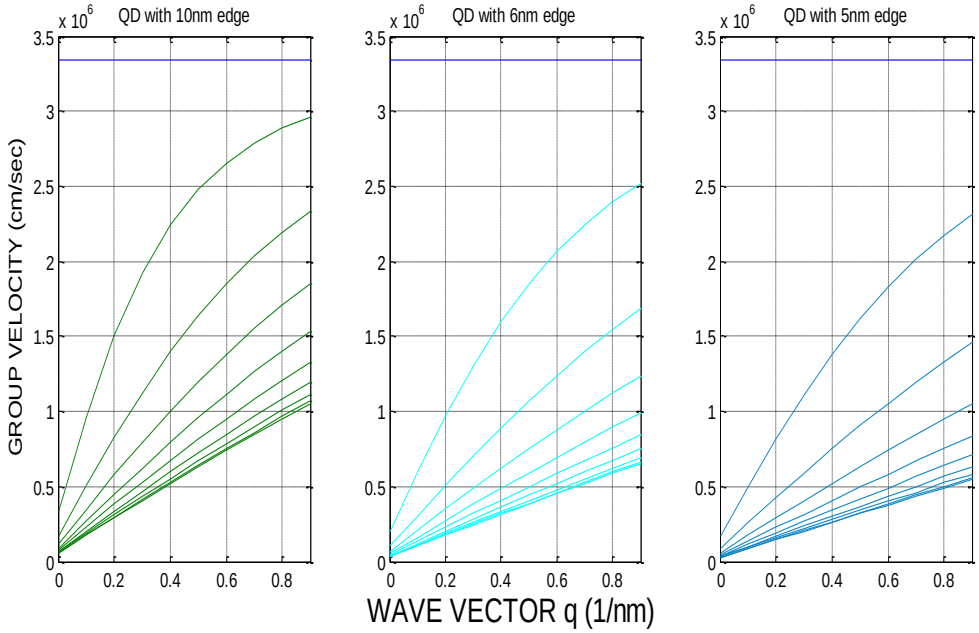

Figure 3. Group Velocity of Quantum Dot with Three Different Sizes [24]

Coating acoustically different material affects the phonon spectra in case of quantum structures [6, 12, 13, 24]. Similarly the phonon spectra of an individual GaAs cubic quantum dot will experience a dramatic change when coated with acoustically dissimilar material. At first we will coat an individual quantum dot with acoustically fast material and observe the phonon spectrum modification. We will choose AlAs as the acoustically fast coating material [22]. We will coat the 6 nm edge GaAs quantum dot with 2 nm AlAs thick layer. Then the total length of the quantum dot edge would be 10 nm as shown in the fig. 4.

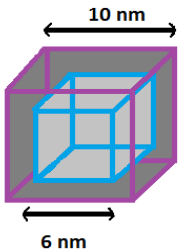

Figure 4. Schematic of a Cubic Quantum dot of GaAs with 6 nm Edge Coated with Acoustically Fast 2 nm AlAs Material

For AlAs elastic stiffness constants $\overline{C_{11}} = 12.02 \times 10^{10}\mathrm{Nm}^{-2}$, $\overline{C_{44}} = 5.89 \times 10^{10}\mathrm{Nm}^{-2}$ and density, $\bar{\rho} = 3598\,\mathrm{Kgm}^{-3}$ [22]. In case of uncoated quantum dot the material parameters was the function of the constituent material for each nodes (10 nodes for 10 equal parts of quantum dot). But in coated quantum dot for the 1st, 2nd, 9th and 10th nodes would take the material parameters of AlAs. The remaining nodes would take the material parameters of GaAs. The eigenvalues, λ are solved and we get the phonon energy. Now we will change the barrier thickness and observe the modification of phonon branches. We will coat the 6 nm edge GaAs quantum dot with 1 nm AlAs thick layer. Then the total length of the quantum dot edge would be 8 nm. Then the phonon spectrum will be modified as shown in the figure 5 which will also be compared with the uncoated one. From this figure we find coating made of material with high sound velocity leads to expansion of phonon branch both qualitatively and quantitatively. For uncoated 6 nm QD 10 branches within 10.2 meV, for coated 10 nm QD 15 branches within 11.859 meV, for coated 8nm QD 15 branches within 12.889 meV. Phonon spectrum is modified differently due to the change in barrier coating. Acoustic impedance mismatch at the interface between the quantum dot and the interface between the quantum dot and the barrier coating affects dramatically the phonon spectra. Since acoustic phonon is modified due to the barrier coating the corresponding group velocity will also be modified as well as other phonon properties. This change of modification will be used for tuning phonon spectrum as we desire.

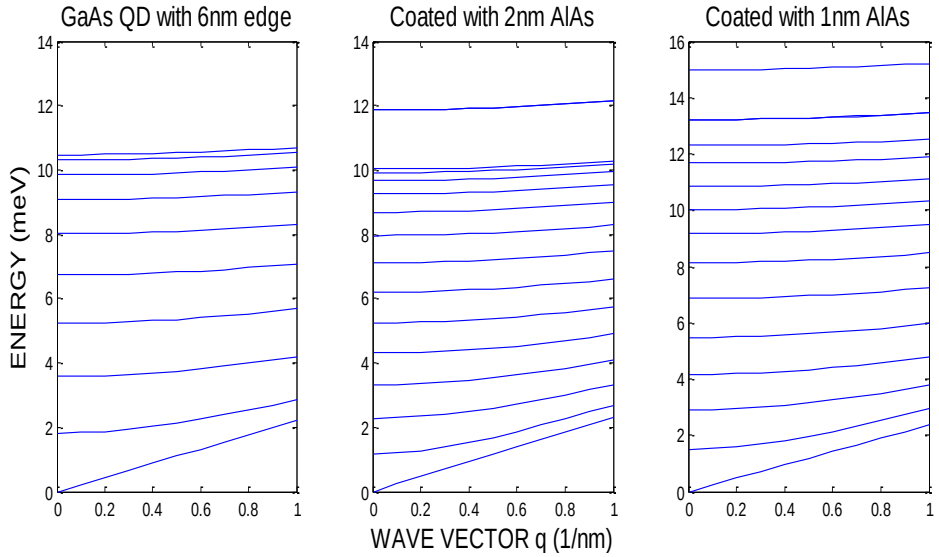

Figure 5. Modification of Phonon Spectrum for Coating with AlAs

Corresponding group velocities are shown in figure 6. Group velocity increases after coating with acoustically fast material AlAs in this case. The first branch which was bulk like in uncoated quantum dot vanishes when quantum dot coated with acoustically dissimilar material. Overall group velocity can be enhanced along certain direction by proper selection of barrier material and its thickness. In case of coated quantum dot with 8 nm edge the group velocity increases more as compared to the coated quantum dot with 10 nm edge. In conclusion we can say that by changing barrier layer thickness we can tune the group velocity as well as other phonon properties such as phonon relaxation rate, phonon density of states phonon scattering etc. Change in Group velocity will modify the lattice thermal conductivity and this process of tuning phonon spectra could be used for phonon engineering for tuning quantum dot.

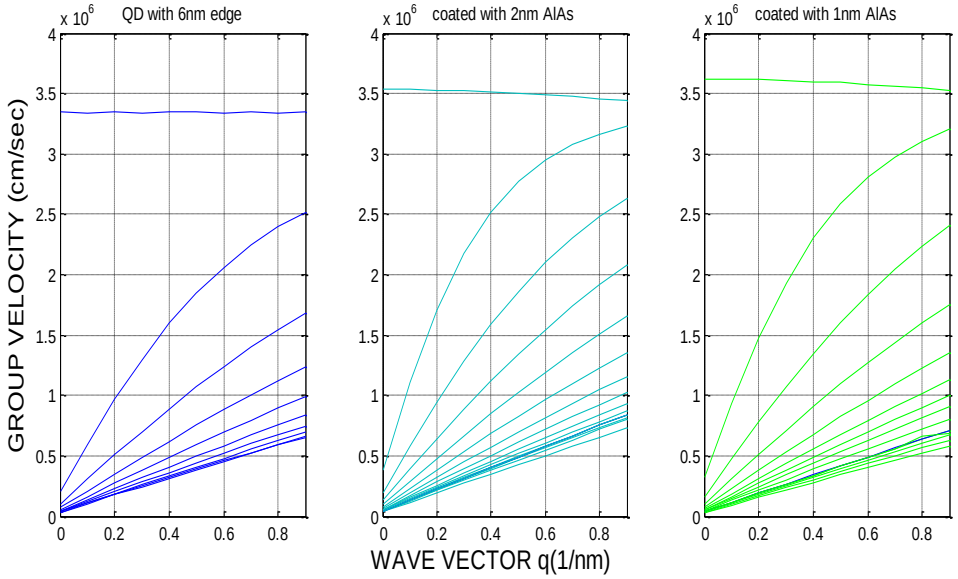

Figure 6. Group Velocity of Coated Quantum Dot Compared with the Uncoated One

We will now choose plastic as the acoustically slow coating material [12, 13]. We will coat the 6 nm edge GaAs quantum dot with 2 nm plastic thick layer. Then the total length of the quantum dot edge would be 10 nm.

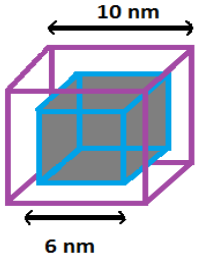

Figure 7. Schematic of a Cubic Quantum Dot of GaAs with 6 nm Edge Coated with Acoustically Slow Plastic Material

For plastic Elastic stiffness constants, $\overline{C_{11}} = 40 \times 10^{10}\mathrm{Nm}^{-2}$ and $\overline{C_{44}} = 10 \times 10^{10}\mathrm{Nm}^{-2}$, Density, $\bar{\rho} = 1000\ \mathrm{Kgm}^{-3}$ [12,13]. Now we observe the phonon spectrum modification of quantum dot coated with acoustically slow material (plastic in this case) which is given in the figure 8.

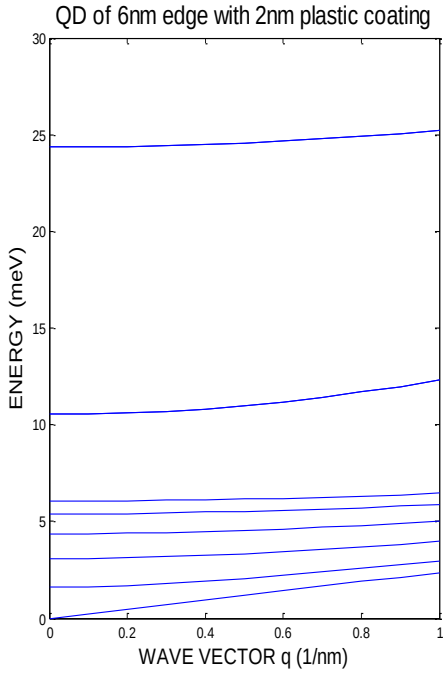

Figure 8. Modification of Phonon Spectrum for Coating with 2 nm Plastic

From this figure we observe that there are changes in phonon dispersion while coating with acoustically slow material. We get only 8 phonon branches in the case of acoustically slow material where lower phonon modes are compressed and two distinct high energy phonon branches with high phononic bandgap i.e. a large phonon stopband is formed which also found theoretically and experimentally for Si and Ge in case of quantum dot crystal [17]. This type of phonon stopband can be used as phonon filter to remove the large number of phonon modes from the thermal transport. Balandin predicted the same thing in his work [1, 20].

4. Conclusion

Strong modification of acoustic phonon dispersion of a GaAs quantum dot is observed and we can conclude that phonon dispersion can be tuned by coating materials. Acoustically fast material such as AlAs leads to the expansion of phonon branches and we find that the group velocity can be increased along certain direction by proper selection of coating material and its thickness. In case of acoustically slow material such as plastic, coating brings about an interesting affect with the exclusion of phonon modes. Thus semiconductor quantum dot coated with acoustically slow material can be used as effective phonon filter eliminating a significant number of phonon modes from thermal transport. From these calculation and observation, we can conclude that by changing the coating materials and their thickness, we can tune the phonon dispersion. Since acoustic phonons are the dominant heat carriers, obtained results of tuning phonon dispersion and group velocity demonstrate a possibility of tuning thermal conductivity of quantum dot which creates an opportunity of using the quantum dot as thermoelectric device as well as for thermal management of downscaled electronic devices.

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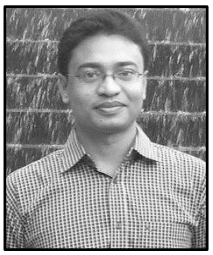

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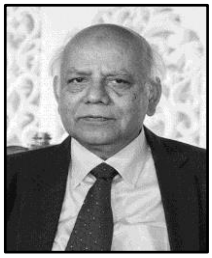

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