THEORY OF PHONON DENSITY OF STATES OF AN ISOTOPICALLY DISORDERED SEMICONDUCTOR CRYSTALS

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ABSTRACT

Semiconductor materials are being widely used in science and technology. The harmonic approximation gives the non-interacting nature of phonons. The concept of harmonicity is never found in nature. In presence of isotopic impurity, translational symmetry of real crystal is destroyed. This creates localized mode. The phonon Green`s function has been taken as containing the entire information of physical property. Fourier transformed phonon Green`s function has been obtained by applying equation of motion technique of quantum dynamics and Dyson equation approach. The response function has been obtained to give the phonon linewidth and phonon shift. Phonon linewidth and phonon shift are further separated into defect and non-defect parts. An expression of phonon density of states (DOS) has been found in terms of diagonal and non-diagonal parts. The effect of temperature, renormalized mode frequency, and perturbed mode frequency on phonon density of states of an isotopically disordered semiconductor crystals has been undertaken in this approach. It is found that excitations are responsible for measure the strength of DOS in both diagonal and non-diagonal parts. It is also found that renormalized mode frequency has been contributed by mass change parameter, force constant change parameter, and electron phonon coupling constant while perturbed mode frequency has been greatly affected by phonon shift.

Keywords: Phonon, Phonon Green`s function, Phonon linewidth, Perturbed mode frequency, Semiconductor, Phonon density of states.

List of notations used in various equations:

1. $\rightarrow$ = Arrow
2. ε = epsilon
3. ω = omega
4. $\hbar$ = hcross

5. $\sum$ = summation

6. θ = theta

7. η = eta

8. Γ = gamma

9. Δ = delta

10. $\bar{\varepsilon}_k$ = bar over ε_k

11. $\tilde{\varepsilon}_k$ = cap over ε_k

12. $\delta_{kk`}$ = Kronecker delta function

13. $\delta(\ )$ = Dirac delta function

14. $\tilde{n}_k$ = cap over n_k

1.Introduction

Technology is being continuously advanced due to large amount of work carried out in semiconductor physics. Semiconductors are the materials whose resistivities are in between metals and insulators. Thermodynamic analysis of ZnO crystal growth from the melt (Asadian, 2013),first - principle studies of phonons III-N compound semiconductors in wurtzite structure (Zhang et al., 2013), microwave-assistant route to hybrid semiconductor nanocrystals with quasi solution - solid - solid mechanism (Bao et al., 2014), semiconductor ZnO nano - rods thin film growth on silver wire for Hemoglobin biosensor fabrication(Battisha et al., 2015) etc. are the latest development in the field of semiconductor technology. It is known that pure crystals are difficult to grow in laboratory. So, real semiconductor crystal can be taken as electron, phonon, and isotopic impurity.These are the basic constituents which are responsible to decide any dynamical characteristics of crystal. The potential energy of a lattice ion is expanded in powers of the displacements of the ions from their equilibrium positions. This expansion upto quadratic term gives the harmonic approximation (Madelung, 1978). This can not explain the complete physical properties such as neutron scattering, Raman scattering, thermal conductivity, phonon lifetime, optical absorption etc. In perfect crystal, wave is generated in the form of normal mode of vibrations having infinite life time. The quantized state of normal mode of vibration is known as phonon. Normally, in real crystals harmonicity breaks due to presence of impurity. This causes change in frequency spectrum of host crystal. This results in a formation of localized mode (Indu,1990). An interaction of electron with this mode gives the formation of dynamical system. According to Hall et al. electron can absorb a photon by indirect transition to conserve the energy and momentum laws at the same time (Kittel,1963). The work on DOS has been done on anharmonic crystal (Indu,1990) but in present case electron phonon interaction is taken with isotopic impurity in low temperature limit for

semiconductor crystals. Theory on the basis of diagonal and non-diagonal parts can be achieved on taking Hamiltonian as a perturbation. This Hamiltonian is contributed by harmonic phonon, electron, electron phonon interaction, and impurity. The defect Hamiltonian has been taken in terms of mass change parameter and force constant change parameter. The phonon density of states is to be developed by dividing this paper into sections namely formulation of the problem, evaluation of Fourier transformed phonon Green`s function via electron phonon defect Hamiltonian, evaluation of phonon density of states and conclusion of the work at the end.

2. Formulation of the problem

The Fourier transformed phonon Green`s function $G_{kk`}(\varepsilon)$ is connected through density of states (DOS) in Lehman representation (Dederichs,1977) as

$$N_{kk`}(\varepsilon) = -\sum_{k} \mathrm{Im}.\, G_{kk`}(\varepsilon) \quad (1)$$

In above eq.(1), $\mathrm{Im}.G_{kk`}(\varepsilon)$ represents imaginary part of phonon Green`s function .The discussion about frequency spectrum will be carried out by the evaluation of the Fourier transformed phonon Green`s function $G_{kk`}(\varepsilon)$. This Green`s function will be obtained by the methodology of equation of motion technique of quantum dynamics.

3. Evaluation of Fourier Transformed Phonon Green`s function Via Electron Phonon Defect Hamiltonian H

The study is done on the basis of phonon Green`s function which is given as (Zubarev D. N., 1960)

$$G_{kk`}(t,t`) = \langle\langle\ A_k(t); A_{k`}^{*}(t`)\ \rangle\rangle = -i\theta(t-t`)[\langle\ A_k(t), A_{k`}(t`)\ \rangle] \quad (2)$$

where $\theta(t-t`)$ is the Heaviside unit step function.

To apply equation of motion technique, first we have to consider the appropriate Hamiltonian. Let us take the following Hamiltonian H as (Maradudin et al., 1971: Frohlich, 1966 : Fan, 1987 : Feinberg et al., 1990 : Mahanty and Behera, 1983 : Behera and Mishra, 1985 : Ziman, 1969 : Sharma and Bahadur, 1975: Sahu and Sharma, 1983: Indu,1990)

$$H = H_{op} + H_{oe} + H_D + H_{ep} \quad (3)$$

The different contributions of Hamiltonian in eq.(2) are H_{op} , H_{oe} , H_{ep} , H_D which represent harmonic part , electron part , electron phonon interaction part , and defect part respectively . These are given in the following equations as

$$H_{op} = \left(\hbar/4\right)\sum_{k}\varepsilon_k\left[A_k^{*}A_k + B_k^{*}B_k\right] \quad (4a)$$

$$H_{oe} = \hbar\sum_{q}\varepsilon_q b_q^{*}b_q \quad (4b)$$

$$H_{ep} = g\hbar\sum_{k,q} b_Q^{*}b_q B_k \quad (4c)$$

$$H_D = -\hbar\sum_{k_1,k_2}\left[C\left(\vec{k}_1,\vec{k}_2\right)B_{k_1}B_{k_2} - D\left(\vec{k}_1,\vec{k}_2\right)A_{k_1}A_{k_2}\right] \quad (4d)$$

The various symbols $b_q^{*}\left(b_q\right), a_k^{*}\left(a_k\right), \varepsilon_k, \varepsilon_q, g, C\left(\vec{k}_1,\vec{k}_2\right)$ and $D\left(\vec{k}_1,\vec{k}_2\right)$ of eqs.(4a-4d) are known as creation(annihilation) operators of electron, creation(annihilation) operators of phonon, phonon frequency(in energy units), electron band energy , electron phonon coupling constant, mass change parameter and force constant change parameter respectively(Maradudin et al., 1971 : Frohlich, 1966 : Fan, 1987 : Feinberg et al.,1990 : Mahanty and Behera, 1983 : Behera and Mishra,1985 : Ziman,1969 : Sharma and Bahadur, 1975: Sahu and Sharma, 1983: Indu,1990).

The Fourier transformed phonon Green`s function is obtained under equation of motion technique of quantum dynamics and Dyson equation approach as(Gairola, 2009 : Indu, 1990 : Painuli et al., 1991 : Sahu and Sharma, 1983 : Sharma and Bahadur, 1975 : Zubarev,1960)

$$G_{kk`}(\varepsilon) = \left(\varepsilon_k\eta_{kk`}/\pi\right)\left[\varepsilon^2 - \bar{\varepsilon}_k^{\,2} + 2i\varepsilon_k\Gamma(kq,\varepsilon)\right]^{-1} \quad (5)$$

In above eq.(5), $\bar{\varepsilon}_k$, $\Gamma(kq,\varepsilon)$ are perturbed mode frequency and phonon linewidth respectively.

The various symbols of eq.(5) are given as

$$\eta_{kk`} = \delta_{kk`} + 4C\left(-\vec{k},\vec{k}`\right)\varepsilon_k^{-1} \quad (6a)$$

$$\bar{\varepsilon}_k^{\,2} = \tilde{\varepsilon}_k^{\,2} + 2\varepsilon_k\Delta(kq,\varepsilon) \quad (6b)$$

$$\tilde{\varepsilon}_k^{\,2} = \varepsilon_k^{\,2} + (2\pi)^{-1}\varepsilon_k\left\{\left\langle\left[M_{kq}(t), B_{k`}^{*}(t`)\right]^0\right\rangle + \left\langle\left[M_{kq}(t), A_{k`}^{*}(t`)\right]^0\right\rangle + 4\varepsilon_k^{-1}\sum_{k_1}C\left(\vec{k},-\vec{k}_1\right)\right.$$

$$\left.\times\left\langle\left[M_{kq}(t), B_{k`}^{*}(t`)\right]^0\right\rangle + 8\left(\varepsilon^2 - \varepsilon_k^{\,2}\right)\varepsilon_k^{-1}C\left(-\vec{k},\vec{k}\right) - 2g\varepsilon_k^{-1}\sum_{q}\left\langle\left[M_{kq}(t), b_Q^{*}b_q\right]^0\right\rangle\right\}_{t=t`} \quad (6c)$$

with,

$$M_{kq}(t) = 4\pi\sum_{k_1}R\left(-\vec{k},\vec{k}_1\right)A_{k_1} - 2\pi L_{kq}(t) \quad (7a)$$

$$R\left(\vec{k}_1,\vec{k}_2\right) = \left(\varepsilon_{k_2}/\varepsilon_{k_1}\right)C\left(\vec{k}_1,\vec{k}_2\right) + D\left(\vec{k}_1,\vec{k}_2\right) + 4\sum_{k_2`}C\left(\vec{k}_1,\vec{k}_2`\right)D\left(-\vec{k}_2`,\vec{k}_2\right)\varepsilon_{k_1}^{-1} \quad (7b)$$

$$L_{kq}(t) = g\sum_{q}\left[\varepsilon_q\left(b_q^{*}b_q + b_Q^{*}b_q\right) + g\left(b_q^{*}b_q B_k + b_Q^{*}b_q B_k\right)\right] \quad (7c)$$

In eq.(6b), $\tilde{\varepsilon}_k$ and $\Delta(kq,\varepsilon)$ are renormalized mode frequency and phonon frequency shift respectively . The response function $P(kq,\varepsilon)$ is found from this approach as

$$P(kq,\varepsilon) = P^D(k,\varepsilon) + P^{(1)}(kq,\varepsilon) + P^{(2)}(kq,\varepsilon) \quad (8)$$

$$P^D(k,\varepsilon) = 8\sum_{k_1} R(\vec{k}_1,-\vec{k}) R^*(\vec{k}_1,-\vec{k}) \varepsilon_{k_1} \left(\varepsilon^2 - \tilde{\varepsilon}_{k_1}^{\,2}\right)^{-1} \quad (9a)$$

$$P^{(1)}(kq,\varepsilon) = 4g^2 \left(N_q + N_Q \delta_{QQ'}\right) \varepsilon_q^{\,2} \left(\varepsilon - 2\varepsilon_q\right)^{-1} \quad (9b)$$

$$P^{(2)}(kq,\varepsilon) = 4g^4 \left(N_q + N_Q \delta_{QQ'}\right) \tilde{n}_k \delta_{kk'} \left(\varepsilon - 2\varepsilon_q\right)^{-1} \quad (9c)$$

where,

$$N_q = \langle b_q^{\,*} b_q \rangle \ , \ N_Q = \langle b_Q^{\,*} b_Q \rangle \ , \ \tilde{n}_k = \langle B_k^{\,*} B_k \rangle \ , \quad (10)$$

The response function $P(kq,\varepsilon)$ can be written in the following form as

$$P(kq,\varepsilon + i\omega) = \Delta(kq,\varepsilon) - i\Gamma(kq,\varepsilon) \quad , \quad \omega \to 0^+ \quad (11)$$

Phonon frequency shift $\Delta(kq,\varepsilon)$ is given by following equation as

$$\Delta(kq,\varepsilon) = \Delta^D(k,\varepsilon) + \Delta^{(1)}(kq,\varepsilon) + \Delta^{(2)}(kq,\varepsilon) \quad (12)$$

where,

$$\Delta^D(k,\varepsilon) = P8\sum_{k_1} R(\vec{k}_1,-\vec{k}) R^*(\vec{k}_1,-\vec{k}) \varepsilon_{k_1} \left(\varepsilon^2 - \tilde{\varepsilon}_{k_1}^{\,2}\right)^{-1} \quad (13a)$$

$$\Delta^{(1)}(kq,\varepsilon) = P4g^2 \left(N_q + N_Q \delta_{QQ'}\right) \varepsilon_q^{\,2} \left(\varepsilon - 2\varepsilon_q\right)^{-1} \quad (13b)$$

$$\Delta^{(2)}(kq,\varepsilon) = P4g^4 \left(N_q + N_Q \delta_{QQ'}\right) \tilde{n}_k \delta_{kk'} \left(\varepsilon - 2\varepsilon_q\right)^{-1} \quad (13c)$$

In eqs.(13a-13c), P indicates for principal value and phonon linewidth $\Gamma(kq,\varepsilon)$ is given by following equation as

$$\Gamma(kq,\varepsilon) = \Gamma^D(k,\varepsilon) + \Gamma^{(1)}(kq,\varepsilon) + \Gamma^{(2)}(kq,\varepsilon) \quad (14)$$

$$\Gamma^D(k,\varepsilon) = 8\pi\omega(\varepsilon)\sum_{k_1} R(\vec{k}_1,-\vec{k}) R^*(\vec{k}_1,-\vec{k}) \varepsilon_{k_1} \delta\left(\varepsilon^2 - \tilde{\varepsilon}_{k_1}^{\,2}\right) \quad (15a)$$

$$\Gamma^{(1)}(kq,\varepsilon) = 4g^2 \left(N_q + N_Q \delta_{QQ'}\right) \varepsilon_q^{\,2} \ \delta\left(\varepsilon - 2\varepsilon_q\right) \quad (15b)$$

$$\Gamma^{(2)}(kq,\varepsilon) = 4g^4 \left(N_q + N_Q \delta_{QQ'}\right) \tilde{n}_k \delta_{kk'} \delta\left(\varepsilon - 2\varepsilon_q\right) \quad (15c)$$

$\tilde{n}_k$ can be obtained with the help of Fourier transformed phonon Green`s function eq.(5) and phonon linewidth eq.(15b) as(Pathak,1965)

$$\tilde{n}_k = 32g^2\hbar^{-1}k_B T \varepsilon_k \varepsilon_q^{\,3} \left(N_q + N_Q \delta_{QQ'}\right) \eta_{kk'} \left(4\varepsilon_q^{\,2} - \bar{\varepsilon}_k^{\,2}\right)^{-2} \quad (16)$$

Eq.(15c) is modified with the help of eq.(16) as

$$\Gamma^{(2)}(kq,\varepsilon) = 128 g^6 \hbar^{-1} k_B T \varepsilon_k \varepsilon_q^3 \delta_{kk`} \left(N_q + N_Q \delta_{QQ`}\right)^2 \eta_{kk`} \left(4\varepsilon_q^2 - \bar{\varepsilon}_k^2\right)^{-2} \delta(\varepsilon - 2\varepsilon_q) \quad (17)$$

4.Evaluation of Phonon Density of States

The phonon density of states is evaluated by substituting eq.(5) into eq.(1) as(Indu,1990)

$$N(\varepsilon) = 2\pi^{-1} \sum_k \varepsilon_k^2 \eta_{kk`} \Gamma(kq,\varepsilon) \left[\left(\varepsilon^2 - \bar{\varepsilon}_k^2\right)^2 + 4\varepsilon_k^2 \Gamma^2(kq,\varepsilon)\right]^{-1} \quad (18)$$

If $\Gamma(kq,\varepsilon)$ is small negligible, Eq.(18) can be taken as

$$N(\varepsilon) = 2\pi^{-1} \sum_k \varepsilon_k^2 \eta_{kk`} \Gamma(kq,\varepsilon) \left(\varepsilon^2 - \bar{\varepsilon}_k^2\right)^{-2} \quad (19)$$

The phonon density of states (DOS) eq.(19), after substituting eq.(14) through eqs.(15a,15b,

17) , can be solved as(Indu,1990)

$$N(\varepsilon) = N_d^D(\varepsilon) + N_{nd}^D(\varepsilon) + N_d^{(1)}(kq,\varepsilon) + N_{nd}^{(1)}(kq) + N_d^{(2)}(kq) + N_{nd}^{(2)}(kq) \quad (20)$$

where,

$$N_d^D(kq) = 32 \sum_k \varepsilon_k^2 R(\vec{k}_1, -\vec{k}) R^*(\vec{k}_1, -\vec{k}) \varepsilon_{k_1} \tilde{\varepsilon}_{k_1} \bar{\varepsilon}_k^2 \left(\tilde{\varepsilon}_{k_1}^4 - \bar{\varepsilon}_k^4\right)^{-2} \quad (21a)$$

$$N_{nd}^D(kq) = 128 \sum_k C(-\vec{k}, \vec{k}`) R(\vec{k}_1, -\vec{k}) R^*(\vec{k}_1, -\vec{k}) \varepsilon_k \varepsilon_{k_1} \tilde{\varepsilon}_{k_1} \bar{\varepsilon}_k^2 \left(\tilde{\varepsilon}_{k_1}^4 - \bar{\varepsilon}_k^4\right)^{-2} \quad (21b)$$

$$N_d^{(1)}(kq) = 8 g^2 \varepsilon_k^2 \varepsilon_q^2 \left(N_q + N_Q \delta_{QQ`}\right) \left(4\varepsilon_q^2 - \bar{\varepsilon}_k^2\right)^{-2} \quad (21c)$$

$$N_{nd}^{(1)}(kq) = 32 C(-\vec{k}, \vec{k}`) g^2 \varepsilon_k \varepsilon_q^2 \left(N_q + N_Q \delta_{QQ`}\right) \left(4\varepsilon_q^2 - \bar{\varepsilon}_k^2\right)^{-2} \quad (21d)$$

$$N_d^{(2)}(kq) = 256 g^6 \hbar^{-1} k_B T \varepsilon_k^3 \varepsilon_q^3 \delta_{kk`} \left(N_q + N_Q \delta_{QQ`}\right)^2 \left(4\varepsilon_q^2 - \bar{\varepsilon}_k^2\right)^{-4} \quad (21e)$$

$$N_{nd}^{(2)}(kq) = 512 C(-\vec{k}, \vec{k}`) g^6 \hbar^{-1} k_B T \varepsilon_k^2 \varepsilon_q^3 \delta_{kk`} \left(N_q + N_Q \delta_{QQ`}\right)^2 \left(4\varepsilon_q^2 - \bar{\varepsilon}_k^2\right)^{-4} \quad (21f)$$

In eqs.(21a-21f), d and nd represent the participation due to diagonal and non diagonal terms respectively.

5. Conclusion

It is concluded from the present study that diagonal and non-diagonal parts have successfully explained the phonon DOS through defect part and non-defect part. It is found that mass change parameter is the decided factor for non-diagonal contribution. In absence of mass change parameter, only diagonal part gives the major role to explain the phonon DOS. The imaginary part of Fourier transformed phonon Green`s function responsible for density of states provides the defect and non-defect parts. The defect part is independent on temperature, and electron phonon coupling constant while non-defect part linearly vary with temperature and non- linearly vary with electron phonon coupling constant. The

temperature dependence in non-defect part is due to the strong coupling g^6 of electron and phonon in comparison to weak coupling contribution g^2. An electron propagates in localized phonon field and harmonic field generates renormalized localized mode frequency and two exciton bound state respectively. These excitations are responsible to broaden the delta function peaks and thus, they are responsible for various contributions regarding phonon density of states through phonon linewidth in semiconductor crystals. The phonon DOS behaves asymptotically in the limit renormalized localized mode frequency in defect part and two exciton bound state in non-defect part are identical with perturbed mode frequency. The formation of exciton occupation number and polaron occupation number are the main results obtained from this Green`s function method in low temperature limit. The renormalized localized mode frequency not only depends on mass change parameter and force constant change parameter but also on electron phonon coupling constant. It is also concluded that perturbed mode frequency is affected as non-linearly on electron phonon coupling constant via renormalized mode frequency.

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