Radboud University

Phonon Damping in Graphene

Bachelor Thesis

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1 Introduction

In a periodic crystalline lattice a quantum of vibrational energy, caused by collective oscillations of atoms in the crystal, is called a phonon. Phonons play an essential role in the conductivity of heat and sound, and the optical, elastic and electrical properties in the lattice. An example of such crystalline lattice is graphene, which is a single layer of carbon atoms arranged in a honeycomb pattern, and the first truly observed 2D crystal.
Due to theoretical calculation done by Landau and Peierls a 2D crystal was presumed not to be able to exist since it would not be stable [1]. At finite temperature, the mean-square atomic displacement would violate the condition that this mean square atomic-displacement should be smaller than the lattice spacing and therefore the harmonic approximation could not be applied to 2D crystals [1]. However detailed analysis beyond the harmonic approximation shows that membranes can exist in a stable and microscopic flat state even without external tension or a substrate.
Exceptional electronic properties such as Dirac fermions and the Quantum Hall effect, thermal properties such as a high thermal conductivity dominated by phonon transport at large wavelengths and mechanical properties, make graphene an intensively studied material both experimentally as theoretically [2].

Due to the measured thermal conductivity being in the range of 3000-5000 W/mK at room temperature, which is very high compared to other heat conduction materials, graphene can be potentially used in many thermal applications [2]. In general heat transport is done by charge carriers, such as electrons, and phonons, but in graphene the electronic thermal conductivity is measured to be less than 1 % [4]. Therefore the thermal conductivity is mainly determined by phonons.

Many different values for the thermal conductivity have been found, mainly due to it being size dependent but also due to different defect concentrations and edge roughness of the crystalline membrane, at which phonons can scatter [4]. Therefore determining the phonon lifetime in graphene is important to understand the extraordinary high thermal conductivity in this crystalline membrane.

In this thesis I calculate the scattering rates of long-wavelength acoustic phonons in free-standing two-dimensional graphene at 0K, within a continuum elasticity description. To determine the scattering rates, I derive an expression for the creation and annihilation operators for the transversal, longitudinal and flexural phonons in graphene in section 3. The interaction Hamiltonian which will be derived within the continuum elasticity theory, will be expressed in these phonon creation and annhiliation operators such that the possible phonon scattering processes in graphene can easily been seen. The decay rates of phonon scattering processes will be determined by Fermi's Golden Rule in section 4. These turned out to be proportional to the initial momentum of the in-plane phonon when decaying in two out-of plane phonons. The lifetime of the phonon is proportional to the inverse of the scattering rate, and should be long enough such that the phonon existed long enough to travel one of its wavelengths. By considering the scattering of both one transversal phonon into two flexural phonons and one longitudinal phonon to two out of plane phonons, I determined that the in-plane phonons are well defined in this crystalline membrane at 0K.

1.1 Lattice Structure of Graphene

Six different phonon branches can be found in graphene. To understand this, we need to describe the atomic structure of graphene. Figure 1 shows the hexagonal lattice of graphene, where the distance between two neighbouring carbon atoms is d and the two lattice vectors $\vec{a_1}$ and $\vec{a_2}$. For graphene these lattice vectors have a value of:

$$\begin{aligned} \vec{a_1} &= d\begin{bmatrix} \sqrt{3} \\ 0 \end{bmatrix} \\ \vec{a_2} &= \frac{d}{2}\begin{bmatrix} \sqrt{3} \\ 3 \end{bmatrix} \end{aligned} \tag{1}$$

The unit cell, of which the lattice is built of, is repeated along these lattice vectors. Therefore the crystal lattice is translation invariant. The unit cell of graphene is build of two carbon atoms, and are depicted as blue and red dots in figure 1 .

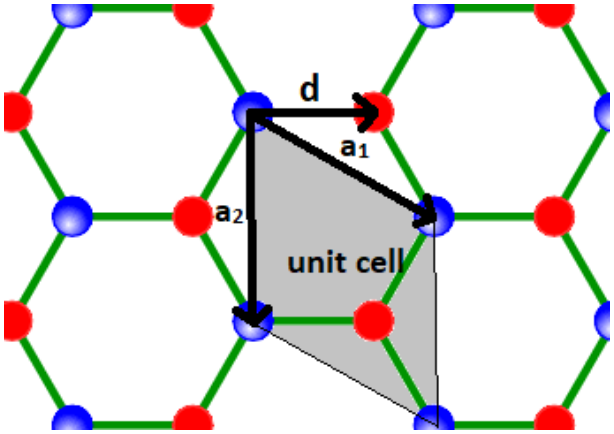

Figure 1: The hexagonal lattice structure of graphene, were the red and blue dots represent the two atoms in the unit cell, a_1 and a_2 the two lattice vectors and d the lattice constant.

Since we are interested in phonons, which can be described as collective movements of the atoms in the crystal, it is convenient to describe the lattice in the reciprocal space. The reciprocal space can be seen as the Fourier transform of the lattice, resulting in a transformation from Cartesian coordinates to a space expressed in wavevectors $\vec{k}$. The reciprocal lattice vectors can be found by using the relation

$$\vec{b_i} \cdot \vec{a_j} = 2\pi\delta_{ij} \tag{2}$$

which then leads to the following reciprocal lattice vectors for graphene:

$$\begin{aligned} \vec{b_1} &= \frac{2\pi}{d\sqrt{3}}\begin{bmatrix} 1 \\ -\frac{1}{\sqrt{3}} \end{bmatrix} \\ \vec{b_2} &= \frac{4\pi}{3d}\begin{bmatrix} 0 \\ 1 \end{bmatrix} \end{aligned} \tag{3}$$

In fig. 2 it is shown that with these two reciprocal lattice vectors the first Brillouin zone of graphene can be depicted. The first Brillouin zone is defined as the Wigner-Seitz primitive cell of the reciprocal lattice, the set of points which are closest to K=0 than other reciprocal lattice point [5]. Since phonon modes differing by a reciprocal lattice vector are equivalent, it is sufficient to look at the first Brillouin zone.
The Γ, K, and M points, representing critical points in the Brillouin zone, are the point having the highest symmetry in graphene and are given by

$$\begin{aligned} \Gamma &= \tfrac{2\pi}{d\sqrt{3}}\begin{bmatrix} 0 \\ 0 \end{bmatrix} \\ K &= \tfrac{2\pi}{d\sqrt{3}}\begin{bmatrix} \frac{2}{3} \\ 0 \end{bmatrix} \\ M &= \tfrac{2\pi}{d\sqrt{3}}\begin{bmatrix} \frac{1}{2} \\ \frac{1}{2\sqrt{3}} \end{bmatrix}. \end{aligned} \tag{4}$$

Following the high symmetry path $\Gamma K M \Gamma$ gives the shaded blue area, which represents the irreducible Brillouin zone in figure 2. Along this path the phonon dispersion relations are often plotted.

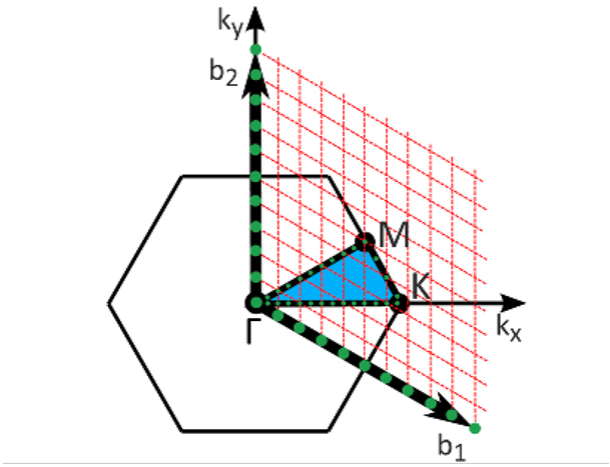

Figure 2: The hexagonal reciprocal lattice structure of graphene, were b_1 and b_2 represent the reciprocal lattice vectors, Γ, K and M the critical points and the blue area the irreducible Brillouin zone [6]

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1.2 Lattice Dynamics and Phonon Branches

The vector corresponding to the coordinates of the atoms can be expressed as:

$$\vec{x}_{nj} = \vec{x}^0_{nj} + \vec{u}_{nj} \tag{5}$$

where n is the label for which unit cell the specific atom is in and j the label for which sublattice [3]. $\vec{x}^0_{nj}$ corresponds to the equillibrium position of the atom and $\vec{u}_{nj}$ the displacement from that position.

If we assume that the mean square atomic displacement is smaller than the lattice spacing between the atoms, see eq. 7, then we can expand the potential energy of the system up to the second order in $\vec{u_{nj}}$ as is done for harmonic oscillators [1]:

$$V(\vec{x}_{nj}) = v(\vec{x}^0_{nj}) + \frac{1}{2} \sum_{n,n',i,j,\alpha,\beta} A^{\alpha\beta}_{ni,n'j} u^{\alpha}_{ni} u^{\beta}_{n'j} \tag{6}$$

with

$$\langle \vec{u_{nj}}^2 \rangle \ll d^2. \tag{7}$$

Here $A^{\alpha\beta}_{ni,n'j}$ is defined as the force-constant matrix, a matrix of second-order partial derivatives of the total potential energy with respect to atomic displacements [1]:

$$A^{\alpha\beta}_{ni,n'j} = (\frac{\partial^2 V}{\partial u^{\alpha}_{ni} \partial u^{\beta}_{n'j}})_{\vec{u}=0} \tag{8}$$

Then the dynamical matrix $D^{\alpha\beta}_{ij}(\vec{k})$ defined as the mass-reduced Fourier transform of the force constant matrix (eq. 9), has eigenvalues which correspond to the squared phonon frequencies[3].

$$D^{\alpha\beta}_{ij}(\vec{k}) = \sum_n \frac{A^{\alpha\beta}_{0i,nj}}{\sqrt{M_i M_j}} \exp(i\vec{k}\vec{x_n}) \tag{9}$$

Here M_i and M_j are the masses of atom i and j, which for graphene both are the mass of a carbon atom, and $\vec{k}$ the wavenumber of the wave describing the phonon. Rigid shifts in the crystal may not result in forces, which is a requirement due to the translational invariance of the crystal and therefore $\sum_{nj} A^{\alpha\beta}_{0i,nj} = 0$, which gives[1]:

$$\sum_j D^{\alpha\beta}_{ij}(\vec{k} = 0) = 0 \tag{10}$$

This results in six phonon branches in graphene, of which a plot can be seen in fig. 3

1. The optical flexural phonon mode, abbreviated as ZO, where at $|\vec{k}| \to 0$ the two atoms in the unit cell move out of plane with a phase difference of π.

$$\omega^2_{ZO}(\vec{k}) = D^{zz}_{11}(\vec{k}) - D^{zz}_{12}(\vec{k}) \qquad (|\vec{k}| \to 0) \tag{11}$$

2 and 3. The two optical in plane modes, abbreviated as TO and LO, again where $\vec{k} = 0$ the movement of the two atoms in the unit cell move out of phase with respect to each other and having squared frequencies equal to the eigenvalue of:

$$\omega^2(\vec{k}) = D^{\alpha\beta}_{11}(\vec{k}) - D^{\alpha\beta}_{12}(\vec{k}) \qquad (|\vec{k}| \to 0) \tag{12}$$

4. The acoustic flexural phonon mode, abbreviated as ZA, where the displacement of the atoms is perpendicular to the surface of the crystalline membrane and without a phase difference

$$\omega^2_{ZA}(\vec{k}) = D^{zz}_{11}(\vec{k}) + D^{zz}_{12}(\vec{k}) \qquad (|\vec{k}| \to 0) \tag{13}$$

5 and 6. Two acoustic phonon modes, where the displacement is in the crystalline membrane plane and where in the limit of $|\vec{k}| \to 0$ the squared frequencies of the transverse and longitudinal modes are determined by the diagonalization of:

$$\omega^2(\vec{k}) = D_{11}^{\alpha\beta}(\vec{k}) + D_{12}^{\alpha\beta}(\vec{k}) \qquad (|\vec{k}| \to 0) \tag{14}$$

Due to eq. 10, it can be shown that the frequencies of all acoustic modes (LA, TA and ZA) vanish in the long-wavelength limit. In fact, it can be shown that for the the in-plane modes $\propto \vec{k}$, while $\omega_{ZA}(\vec{k}) \propto \vec{k}^2$ [11]. This follows from the rotational invariance of the crystalline membrane in 2D. Therefore the acoustic flexural modes are not linear in wavenumber, as the TA and LA phonons are and just as sound waves normally are, but quadratic at the limit of $\vec{k} \to 0$. The frequency of ZA phonons vanishes more quickly at this limit. This ZA phonon branch has important consequences for the thermal properties and the stabilization of graphene [1].

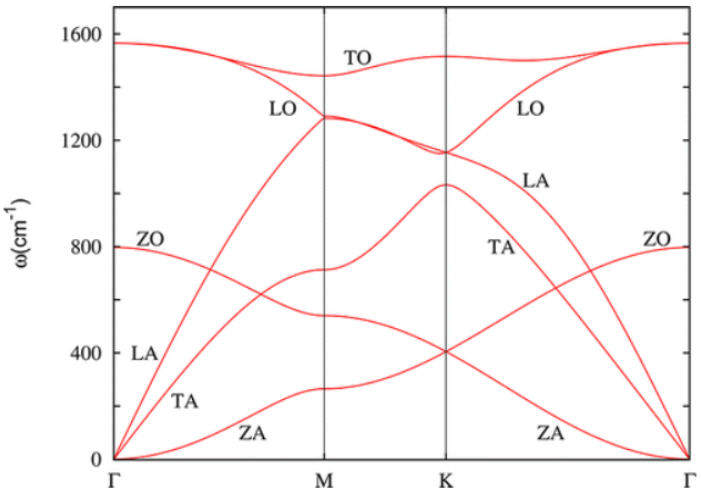

Figure 3: Phonon spectrum of graphene along the high symmetry path [1]

1.3 Elasticity Theory of Flexible Crystalline Membranes

At finite temperature the harmonic approximation is invalid for 2D crystalline membranes due to the violation for both in plane and out of plane acoustic phonons of the condition that can be found in eq. 7 [1]. The anharmonic interactions have to be taken account, which can be formulated by the continuum elasticity theory [1]. In the crystalline membrane, initially in the $\hat{x}\hat{y}$ plane, we now define $u_\alpha(x_\beta)$ as the displacements of the atom, where α and β stand for the $\hat{x}$ or $\hat{y}$ direction and a displacement perpendicular to the membrane as h. Then the distance between two points in the membrane is changed from dl^2 to $dl'^2 = dl^2 + 2u_{\alpha\beta}dx_\alpha dx_\beta$ where $u_{\alpha\beta}$ is the deformation tensor [3]:

$$u_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} + \frac{\partial u_\gamma}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\alpha} + \frac{\partial h}{\partial x_\beta}\frac{\partial h}{\partial x_\alpha}\right). \tag{15}$$

The mechanical forces in the crystal plane, which are in general stress forces are given by[3]:

$$f_\alpha = \oint dS_\beta \sigma_{\alpha\beta}. \tag{16}$$

Here dS_β is a vector element of the crystalline surface and $\sigma_{\alpha\beta}$ is the stress tensor which describes the stress in the lattice membrane as a result of a lattice deformation. For an isotropic elastic membrane the stress tensor can be written as

$$\sigma_{\alpha\beta} = \lambda\delta_{\alpha\beta}u_{\gamma\gamma} + 2\mu u_{\alpha\beta} \tag{17}$$

where λ and μ are Lamé constants, of which the value is material depending [3]. For graphene these values at a temperature of 0K are $\lambda \approx 3.8\frac{eV}{\text{Å}^2}$ and $\mu = 9.3\frac{eV}{\text{Å}^2}$ [10].
This results in a deformation energy of [3]

$$F = \frac{1}{2}\int \left\{\lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta}\right\}\ d^2x. \tag{18}$$

Mechanical forces perpendicular to the crystal surface result in strain, which bends the crystalline membrane. The bending energy is given in equation 19 in which κ is the bending rigidity. For graphene $\kappa \approx 1.1eV$ [3].

$$F_{bending} = \frac{\kappa}{2}\int (\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2\ d^2x. \tag{19}$$

Then the total deformation energy is given by [3]

$$F = \frac{1}{2}\int \left\{\kappa[(\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2 + \lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta}\right\}\ d^2x. \tag{20}$$

The system can be set in a frame, such that an external isotropic tension (σ^{ext}) is applied on the edges of crystalline membrane. This gives an additional term in the total deformation energy, which is then [3].

$$F = \frac{1}{2}\int \left\{\kappa[(\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2 + \lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta} + 2\sigma^{ext}u_{\alpha\alpha}\right\}\ d^2x. \tag{21}$$

1.4 Creation and Annihilation Operators

Due to the fact that phonons describe oscillations, we will start with an introduction of the creation and annihilation operators of the 1D harmonic oscillator. These operators are especially useful in the occupation number representation where the wavefunction is depicted as ket which specifies the number of particles of a sort. For example the wavefunction $|n_i, n_j> = |3, 2>$ which is a state of two types of particles i and j of which 3 particles of type i and 2 of type j are found. This can also be done for particles with different momenta k, such that we can define the creation operator $\hat{a}^\dagger(k_i)$, creating a particle with momentum k_i and annihilation operator $\hat{a}(k_i)$, destructing a particle which has a momentum of k_i. If we are initially in a vacuum state $|n_{k_1}, n_{k_2}, n_{k_3} > = |0, 0, 0>$ or a state without a particle with momentum k_i and apply the annihilation operator $\hat{a}(k_i)$ on this state, this results in zero since their is no particle with momentum k_i that can be annihilated. The same

accounts for a creation operator $\hat{a}^\dagger(k_i)$ acting on a bra-state without a particle with momentum k_i. Because phonons are bosonic particles, we have the following commutation relations which the creation and annihilation operators have to satisfy[7]

$$\begin{aligned}[\hat{a}_j^\dagger, \hat{a}_k^\dagger] = [\hat{a}_j, \hat{a}_k] = 0 \\ [\hat{a}_j, \hat{a}_k^\dagger] = \delta_{jk}\hat{1}.\end{aligned} \tag{22}$$

These we can later use to commute the annihilation operators in front of the ket or the creation operators in front of the bra, such that it is easy seen which expressions are zero.

Now we will derive the creation and annihilation operators for a set of simple 1D harmonic oscillators, which can be easily done by defining the dimensionless operators $\hat{X}(k)$ and $\hat{P}(k)$. The Hamiltonian expressed in the momentum operator $\hat{p}(k)$ and the position operator $\hat{x}(k)$ for the 1D quantum harmonic oscillator is given by[7]:

$$H = \frac{\hat{p}^2(k)}{2m} + \frac{1}{2}m\omega^2\hat{x}^2(k). \tag{23}$$

The momentum operator can be expressed as $\hat{p}(k) = -i\hbar\frac{\partial}{\partial x(k)}$ and where the position and momentum operator satisfy the commutation relation: $[\hat{x}(k), \hat{p}(k')] = i\hbar\delta_{k,k'}$. The dimensionless momentum and position operator are expressed as:

$$\begin{aligned}\hat{P}(k) = \frac{\hat{p}(k)}{\sqrt{m\hbar\omega(k)}} \\ \hat{X}(k) = \hat{x}(k)\sqrt{\frac{m\omega(k)}{\hbar}}\end{aligned} \tag{24}$$

Then the hamiltonian can be written as:

$H = \frac{\hbar\omega(k)}{2}[\hat{P}(k)^2 + \hat{X}(k)^2] = \frac{\hbar\omega(k)}{2}[\hat{P}(k) + i\hat{X}(k)][\hat{P}(-k) - i\hat{X}(-k)] + \frac{\hbar\omega(k)}{2}$

If we introduce the following creation and annihilation operators,

$$\begin{aligned}\hat{a} = \frac{1}{\sqrt{2m\hbar\omega(k)}}\hat{p}(k) - i\sqrt{\frac{m\omega(k)}{2\hbar}}\hat{x}(k) \\ \hat{a}^\dagger = \frac{1}{\sqrt{2m\hbar\omega(k)}}\hat{p}(k) + i\sqrt{\frac{m\omega(k)}{2\hbar}}\hat{x}(k)\end{aligned} \tag{25}$$

the Hamiltonian can be rewritten in the well known form:

$$H = \hbar\omega(k)\left[\hat{a}^\dagger(k)\hat{a}(k) + \frac{1}{2}\right]. \tag{26}$$

Since phonons describe oscillations in the lattice, the structure of the creation and annihilation operators of phonons will be similar, which will be derived in section 3.

1.5 Fermi's Golden Rule

Fermi's Golden Rule can be used to calculate the transition rate of going from an initial state $|i>$ to a distribution of final states $<f|$ when a first order perturbation is being applied. Here the perturbation is time independent. The transition rate is proportional to the coupling strength between the initial and final states of the system which is expressed by $|<f|H_{interaction}|i>|^2$ and to the density of the final states $D(E_{final})$ [8]. To obtain Fermi's golden rule, let us first have a look at the relevant expansion coefficient at first order in perturbation theory. The time evolution of a state is generally expressed as

$$|\psi(t,x)> \sum_k c_k exp(-i\frac{E_k^0 t}{\hbar})|\psi_k(x)> \tag{27}$$

This can be substituted in the time dependent Schrödinger Equation, $i\hbar\frac{\partial}{\partial t}\psi(t,x) = (H_0+H')\Psi(t,x)$. giving:

$\mathrm{i}\hbar\sum_k \dot{C}_k(t)\exp(\frac{-iE_k^0 t}{\hbar})|\Psi_k(x)> + i\hbar\sum_k C_k(t)\frac{-iE_k^0}{\hbar}\exp(\frac{-iE_k^0 t}{\hbar})\Psi_k(x)>$
$= (H_0+H')\sum_k C_k(t)\exp(\frac{-iE_k^0 t}{\hbar})|\Psi_k(x)>$

The second term on the left hand side cancels out the H_0 term on the right. From this the relevant expansion coefficient $c_{fi}(t)$ between the initial state $|i>$ and final state $<f|$, can be derived which then leads to:

$$\begin{aligned} c_{fi}(t) &= -\tfrac{i}{\hbar}H'_{fi}\int_0^t dt' \exp(\tfrac{i(E_i^0-E_f^0)t'}{\hbar}) \\ &= \tfrac{H'_{fi}}{E_f^0-E_i^0}(1-exp(\tfrac{i(E_i^0-E_f^0)}{\hbar})). \end{aligned} \tag{28}$$

Then the probability $P_{fi}(t) = |<f|i>|^2$ of going from initial state $|i>$ to final states $<f|$ can be expressed as the square of the relevant expansion coefficient, resulting in a transition probability of

$$P_{fi}(t) = \int dE_f D(E_f)\frac{4|H'_{fi}|^2}{|E_f^0-E_i^0|^2}\sin^2(\frac{(E_i^0-E_f^0)t}{2\hbar}). \tag{29}$$

where $D(E_f)$ gives the density of the final states. The transition rate $R_{i\to|f|}$is defined as the transition probability per unit time and therefore is equal to the derivative to time of the transition probability in eq. 29. Using the mathematical identity of eq. 74 we get to Fermi's Golden Rule, where $|H_{fi} =< f|H_{int}|i >$:

$$R_{i\to|f|} = \frac{d}{dt}\sum_{final\ states} P_{fi}(t)\delta(E_f-E_i) = \frac{2\pi}{\hbar}\sum_{final\ states}|H_{fi}|^2\delta(E_f-E_i) \tag{30}$$

However the transition probability in Fermi's Golden Rule is not sufficient when transition probabilities are determined for scattering processes that concern intermediate states. Considering perturbation up to the second order, the transition amplitude for going from going to initial state $|i>$ to final states $<f|$, with intermediate state $|m>$ can be written as

$$\sum_{final\ states}\Big(<f|H_{int}|i> + \sum_m \frac{<f|H_{int}|m><m|H_{int}|i>}{E_i-E_m}\Big) \tag{31}$$

With the scattering rate of all possible scattering decays, the lifetime of the phonon can be determined, which is equal to the inverse of the scattering rate. The lifetime of phonons determines, among other properties, the heat conductivity in the crystal. Furthermore we can use this as a measure to determine if the phonon is well defined. A well defined phonon propagates long enough such that it travelled at least a distance equal to one of its wavelength [9]. The time required to travel this distance is $\frac{2\pi}{\omega(k)}$, such that a well defined phonon should satisfy:

$$\frac{\omega(k)}{R} \geq 2\pi \tag{32}$$

2 Phonon Dispersion Relations

As a first step to compute the scattering rates, I expanded the Hamiltonian, specifying the energy of the collective atom oscillations in graphene, in phonon creation and annihilation operators. Therefore, the expression for phonon creation and annihilation needs to derived, which requires a computation of phonon frequencies in graphene. This can be done by constructing the equation of motion of these vibrations and extracting the different frequencies by assuming that the collective vibrations can be described by plane waves. The equation of motion can be obtained by the Euler-Langrange equation.

In the Lagrangian, $\mathcal{L} = T - U$, the kinetic term can be written as

$$T = \int \frac{\rho}{2}((\frac{dh}{dt})^2 + (\frac{du_\alpha}{dt})^2)\ d^2x \tag{33}$$

In which ρ for graphene has a value of $7.6114 \times 10^{-7} kg/m^2$. The potential energy is equivalent to the total deformation energy in equation 21 in which the deformation tensor of eq.15 is substituted, such that:

$$\mathcal{L} = \int \frac{\rho}{2}((\frac{dh}{dt})^2 + (\frac{du_\alpha}{dt})^2) - \frac{1}{2}\ \kappa[(\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2 + \lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta} + \sigma^{ext}u_{\alpha\alpha}\big)\ d^2x$$
$$\text{with } u_{\alpha\beta} = \frac{1}{2}(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} + \frac{\partial u_\gamma}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\alpha} + \frac{\partial h}{\partial x_\beta}\frac{\partial h}{\partial x_\alpha}). \tag{34}$$

Since phonons describe atoms vibrating in a quantized normal mode, we can describe the in-plane and out-of-plane vibrations as plane waves:

$$\begin{aligned} h_{ansatz} &= h_0 \exp(i(\vec{k}\vec{x} - \omega t) \\ u_\alpha &= u_\alpha^0 \exp(i(\vec{k}\vec{x} - \omega t). \end{aligned} \tag{35}$$

This leads to the following phonon dispersion relations:

$$\text{ZA phonons:} \qquad \omega_0 = \sqrt{\frac{\kappa k^4 + \sigma^{ext}k^2}{\rho}} \tag{36}$$

$$\text{LA phonons:} \qquad \omega_L = \sqrt{\frac{\kappa k^4 + (\lambda + 2\mu + \sigma^{ext})k^2}{\rho}} \tag{37}$$

$$\text{TA phonons:} \qquad \omega_T = \sqrt{\frac{\kappa k^4 + (\mu + \sigma^{ext})k^2}{\rho}} \tag{38}$$

In my thesis, I will focus on the case of zero external tension.

In most papers the third, nonlinear term of the deformation tensor is ignored, because the out of plane fluctuations are stronger than the in plane fluctuations [1] and therefore can be ignored according L.D. Landau and E.M. Lifshitz 'as being the second order of smallness' in the deformation

tensor[11]. This makes the equation not fully rotational invariant. This assumption results in the TA and LA phonon dispersion being only linear in wavenumber:

$$\text{LA phonons:} \qquad \omega_L = \sqrt{\frac{(\lambda + 2\mu)k^2}{\rho}} \tag{39}$$

$$\text{TA phonons:} \qquad \omega_T = \sqrt{\frac{\mu k^2}{\rho}} \tag{40}$$

I will use these expressions for the acoustic phonon dispersion relations (eq. 39 and eq. 40) because $\frac{\lambda+2\mu}{\kappa k^2}$ is large for $k \to 0$. An because in the limit $\vec{k} \to 0$, $\frac{\kappa}{\rho}k^4$ goes faster to zero then $\frac{(\lambda+2\mu)}{\rho}k^2$ and therefore the quadratic order can be neglected compared to the linear term between $\vec{k} = [0, \sqrt{\frac{\lambda+2\mu}{\kappa}}]$.

3 Hamiltonian in Creation and Annihilation Operators

The Hamiltonian of the crystalline membrane is obtained by adding the total deformation energy (eq. 20) and the kinetic energy and substituting in the deformation tensor (eq. 15).

$$H = \int \frac{\hat{P}_z(x)^2}{2\rho} + \frac{\hat{P}_\alpha(x)^2}{2\rho} + \frac{1}{2}\left\{\kappa[(\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2 + \lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta}\right\}\, d^2x$$
$$\text{with } u_{\alpha\beta} = \frac{1}{2}(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} + \frac{\partial u_\gamma}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\alpha} + \frac{\partial h}{\partial x_\beta}\frac{\partial h}{\partial x_\alpha}) \tag{41}$$

To obtain our Hamiltonian in creation and annihilation operators, we first Fourier transform the operators $\hat{p}_z(x)$, $\hat{p}_\alpha(x)$,$\hat{h}(x)$ and $\hat{u}_\alpha(x)$ to $\hat{p}_z(k)$, $\hat{p}_\alpha(k)$, $\hat{h}(k)$ and $\hat{u}_\alpha(k)$ and find a definition for the phonon creation and annihilation operators. We define the following Fourier transformations:

$$\hat{p}_z(x) = \frac{1}{\sqrt{A}} \sum_{k=-\infty}^{\infty} \exp(ikx)\, \hat{p}_z(k) \tag{42}$$

$$\hat{p}_\alpha(x) = \frac{1}{\sqrt{A}} \sum_{k=-\infty}^{\infty} \exp(ikx)\, \hat{p}_\alpha(k) \tag{43}$$

$$\hat{h}(x) = \frac{1}{\sqrt{A}} \sum_{k=-\infty}^{\infty} \exp(ikx)\, \hat{h}(k) \tag{44}$$

$$\hat{u_\alpha}(x) = \frac{1}{\sqrt{A}} \sum_{k=-\infty}^{\infty} \exp(ikx)\hat{u}_\alpha(k) \tag{45}$$

Here $\hat{u}_\alpha(k) = [\, t^L_\alpha(k)\hat{Q}^L + t^T_\alpha(k)\hat{Q}^T]$, where $t^T_\alpha(k)$ and $t^L_\alpha(k)$ are, respectively, transverse and longitudinal projection vectors, projecting the transversal and longitudinal part of the wavevector such that they have the following properties:

$$\begin{aligned} k_\alpha t^L_\alpha(k) &= |k| \\ k_\alpha t^T_\alpha(k) &= 0 \end{aligned} \tag{46}$$

$$\begin{aligned} t_\alpha^L(k)t_\alpha^L(k) &= 1 \\ t_\alpha^T(k)t_\alpha^T(k) &= 1 \\ t_\alpha^L(k)t_\alpha^T(k) &= 0 \end{aligned} \tag{47}$$

and

$$\begin{aligned} t_\alpha^L(k)t_\beta^L(k) = P_{\alpha\beta}^L(k) &= \tfrac{k_\alpha k_\beta}{k^2} \\ t_\alpha^T(k)t_\beta^T(k) = P_{\alpha\beta}^T(k) &= \delta_{\alpha\beta} - P_{\alpha\beta}^L(k) \end{aligned} \tag{48}$$

By using the orthogonality relation(eq. 49) the number of independent wavenumbers k can be reduced, since this relation implies that only momentum-conserving interactions appear in the Hamiltonian.

$$\int d^2x \ \frac{exp(i(k+k')x}{A} = \delta_{k,-k'} \tag{49}$$

The Hamiltonian can be splitted in a propagation part $H_{propagation}$ quadratic in the displacements h and u_α and depending on one k value due to the orthogonality relation, and a part depending on different k values and therefore describing the interactions between phonons, which we called $H_{interaction}$. $H_{propagation}$ can be obtained by substituting only the first two terms of the deformation tensor in the Hamiltonian, while the $H_{interaction}$ is obtained by the additional terms arising when the full deformation tensor is substituted.

$$\begin{aligned} H = &\int \frac{\hat{P}_z(x)^2}{2\rho} + \frac{\hat{P}_\alpha(x)^2}{2\rho} + \frac{1}{2}\left\{\kappa[(\nabla^2 h)^2 + (\nabla^2 u_\gamma)^2 + \lambda(u_{\alpha\alpha})^2 + 2\mu u_{\alpha\beta}u_{\alpha\beta}\right\} \ d^2x \\ &\text{with } \bar{u}_{\alpha\beta} = \frac{1}{2}(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha}) \text{ leading to } H_{propagation} \\ &\text{and the additional terms when the full deformation tensor is substituted} \\ &u_{\alpha\beta} = \frac{1}{2}(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} + \frac{\partial u_\gamma}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\alpha} + \frac{\partial h}{\partial x_\beta}\frac{\partial h}{\partial x_\alpha}) \text{ leads to } H_{interaction} \ . \end{aligned} \tag{50}$$

Form the propagation part, we can define the creation and annihilation operators of our system as is done for harmonic oscillators in section 1.4. $H_{propagation}$ can be split in a part describing the in plane displacements and a part describing the out of plane displacements, such that we can define phonon specific creation and annihilation operators.

$$H_{propagation} = H_{in\ plane} + H_{out\ plane} \tag{51}$$

$$H_{out\ plane} = \sum_{k=-\infty}^{\infty} \frac{\hat{p}_z(k)\hat{p}_z(-k)}{2\rho} + \frac{\kappa}{2}k^4\hat{h}(k)\hat{h}(-k) \tag{52}$$

$$\begin{aligned} H_{in\ plane} = \textstyle\sum_{k=-\infty}^{\infty} &\tfrac{\hat{p}_\alpha(k)\hat{p}_\alpha(-k)}{2\rho} + \tfrac{\kappa k^4}{2}u_\gamma(k)u_\gamma(-k) + \tfrac{\lambda+\mu}{2}k_\alpha k_\beta u_\alpha(k)u_\beta(-k) \\ &+\tfrac{\mu}{2}k^2 u_\beta(k)u_\beta(-k) \end{aligned} \tag{53}$$

By using the frequencies found in equations 36, 37 and 38 and using the commutation relations $[\hat{h}(k), \hat{p}_z(-k)] = i\hbar\delta_{k,-k}$, $[\hat{u}_\alpha(k), \hat{p}_\beta(-k)] = i\hbar\delta_{\alpha\beta}\delta_{k,-k}$ and the commutation relations for creation

and annihilation operators in equation 22, we find the following expressions for $\hat{p}_z(k)$, $\hat{p}_\alpha(k)$, $\hat{h}(k)$ and $\hat{u}_\alpha(k)$ in phonon creation and annihilation operators:

$$\begin{aligned}
p_z(k) &= -\sqrt{\tfrac{\hbar\rho\omega_0(k)}{2}}[\hat{a}(k)+\hat{a}^\dagger(-k)] \\
\hat{p}_\alpha(k) &= -\sqrt{\tfrac{\hbar\rho\omega_L(k)}{2}}t_\alpha^L(k)[\hat{c}(k)-\hat{c}^\dagger(-k)] - \sqrt{\tfrac{\hbar\rho\omega_T(k)}{2}}t_\alpha^T(k)[\hat{b}(k)-\hat{b}^\dagger(-k)] \\
\hat{h}(k) &= \tfrac{1}{i}\sqrt{\tfrac{\hbar}{2\rho\omega_0(k)}}[\hat{a}(k)-\hat{a}^\dagger(-k)] \\
\hat{u}_\alpha(k) &= \tfrac{1}{i}[t_\alpha^L(k)\sqrt{\tfrac{\hbar}{2\rho\omega_L(k)}}[\hat{c}(k)+\hat{c}^\dagger(-k)] + t_\alpha^T(k)\sqrt{\tfrac{\hbar}{2\rho\omega_T(k)}}[\hat{b}(k)+\hat{b}^\dagger(-k)]
\end{aligned} \tag{54}$$

The creation and annihilation operators $\hat{a}^\dagger(k)$and $\hat{a}(k)$ create or annihilate a ZA phonon, $\hat{c}^\dagger(k)$and $\hat{c}(k)$ create or annihilate a LA phonon, and $\hat{b}^\dagger(k)$and $\hat{b}(k)$ create or annihilate a TA phonon of momentum $\hbar\vec{k}$.

Then the part of the Hamiltonian describing the displacement of the atoms in the lattice can be written as:

$$H_{propagation} = \sum_k \hbar\omega_0(k)\left(\hat{a}^\dagger(k)\hat{a}(k)+\frac{1}{2}\right) + \hbar\omega_L(k)\left(\hat{c}^\dagger(k)\hat{c}(k)+\frac{1}{2}\right) + \hbar\omega_T(k)\left(\hat{b}^\dagger(k)\hat{b}(k)+\frac{1}{2}\right) \tag{55}$$

By substituting the relation for the momentum and position operators expressed in creation and annihilation operators (eq. 54) in the interaction Hamiltonian (eq. 50), it can easily been seen which phonon scattering processes can occur.

$$\begin{aligned}
H_{interaction} = \sum_{k,k',k''} &\frac{\lambda+2\mu}{32A}\frac{\hbar^2}{\rho^2\sqrt{\omega_0(k)\omega_0(k')\omega_0(k'')\omega_0(-k-k'-k'')}}\vec{k}_\alpha\vec{k'}_\alpha\vec{k''}_\beta(-\vec{k}_\beta-\vec{k'}_\beta-\vec{k''}_\beta) \\
&\left(\hat{a}(k)-\hat{a}^\dagger(-k)\right)\left(\hat{a}(k')-\hat{a}^\dagger(-k')\right)\left(\hat{a}(k'')-\hat{a}^\dagger(-k'')\right)\left(\hat{a}(-k-k'-k'')-\hat{a}^\dagger(k+k'+k'')\right)
\end{aligned} \tag{56a}$$

$$\begin{aligned}
&-\frac{\lambda}{4\sqrt{2A}}\frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k)\omega_0(k')\omega_L(-k-k')}}(\vec{k}_\alpha+\vec{k'}_\alpha)\vec{k}_\beta\vec{k'}_\beta \\
&\left(\hat{a}(k)-\hat{a}^\dagger(-k)\right)\left(\hat{a}(k')-\hat{a}^\dagger(-k')\right)\left(t_\alpha^L(-k-k')(\hat{c}_{-k-k'}+\hat{c}^\dagger_{k+k'})\right)
\end{aligned} \tag{56b}$$

$$\begin{aligned}
&-\frac{\mu}{2\sqrt{2A}}\frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k)\omega_0(k')}}(\vec{k}_\alpha+\vec{k'}_\alpha)\vec{k}_\alpha\vec{k'}_\beta)\left(\hat{a}(k)-\hat{a}^\dagger(-k)\right)\left(\hat{a}(k')-\hat{a}^\dagger(-k')\right) \\
&\left[\frac{t_\beta^L(-k-k')}{\sqrt{\omega_L(-k-k')}}(\hat{c}(-k-k')+\hat{c}^\dagger(k+k')) + \frac{t_\beta^T(-k-k')}{\sqrt{\omega_T(-k-k')}}(\hat{b}(-k-k')+\hat{b}^\dagger(k+k')\right]
\end{aligned} \tag{56c}$$

The full interaction Hamiltonian, obtained by substituting the deformation tensor is determined, however for simplicity I only display here the terms that I used in the calculations. The terms

not displayed here, came from the nonlinear term in u in the deformation tensor, $\frac{\partial u_\gamma}{\partial x_\beta}\frac{\partial u_\gamma}{\partial x_\alpha}$, which is often neglected as was mentioned in section 2. These extra terms made more decay processes possible at a lower order interaction than the terms in 56.

Furthermore due to terms containing $t^T_\alpha(k)k_\alpha$, which is equal to zero, the Hamiltonian contains more scattering terms for the longitudinal acoustic phonons than for the transversal acoustic phonons.

4 Scattering Processes

With the interaction Hamiltonian expressed in creation and annihilation operators the possible scattering processes can easily been seen. In this section the scattering between LA phonons and TA phonons will be compared and the computation of the rate of one ZA phonon to three ZA phonon will be explained.

4.1 Scattering of LA and TA phonons

This section presents a calculation of scattering rates of in-plane LA and TA phonons into two ZA phonons. For both scattering processes the transition probability is calculated by
P=$| < f|H_{interaction}|i > |^2$. For $1TA \rightarrow 2ZA$ we define as initial state $|i >= \hat{b}^\dagger(k_1)|0\rangle$, for $1TA \rightarrow 2ZA$ we define $|i >= \hat{c}^\dagger(k_1)|0\rangle$ and for both processes the final state is $< f| =< 0|\hat{a}(k_2)\hat{a}(k_3)$.

The terms in the interaction Hamiltonian (eq. 56) contributing to this probability are the terms with an equal number of the same sort of phonon creation and annihilation operators. Otherwise the $< bra|$ and $|ket >$ will not be equal and the amplitude this term will be zero according the orthonormality relation:

$$< \Psi_1|\Psi_2 >= \delta_{\Psi_1,\Psi_2}. \tag{57}$$

When solving this probability, the commutation relations of the creation and annihilation operators are used, which can be found in 22. In general, the only physical contribution to the scattering rates arise from the terms in which all 'external' operators, $\hat{c}^\dagger(k_1)$, $\hat{b}^\dagger(k_1)$, $\hat{a}(k_2)$ and $\hat{a}(k_3)$ are commuted with 'internal' operators arising in the expansion of the interaction. Processes in which 'external' operators are commuted among themselves describe, instead, phonons which do not participate to interaction processes. However in this process this is not a concern since the initial creation operators $\hat{b}^\dagger(k_1)$ and $\hat{c}^\dagger(k_1)$ are different types of phonon operators than the final phonon operators $\hat{a}(k_2)$ and $\hat{a}(k_3)$.

Then the determined transition probability can be substituted in Fermi's Golden Rule (eq. 30) such that the scattering rate of this process can be determined. By considering momentum conservation, we can limit the number of degrees of freedom. We define our axis such that $\vec{k_1} = (k_1, 0)$, and from momentum conservation it follows that $k_{3x} = k_1 - k_{2x}$ and $k_{3y} = -k_{2y}$. By replacing the summation with a continuum integration and by using the properties of the δ function (Appendix eq. 75 and 76), the scattering rate can be recast as:

$$\begin{aligned} R_{i\rightarrow|f|} &= \frac{d}{dt} \sum_{final\ \ states} P_{fi}(t)\delta(E_f - E_i) \\ &= \frac{2\pi A}{\hbar} \int \frac{\partial k_{2x}}{(2\pi)^2} \frac{P_{i\rightarrow|f|}(+|k_{2y}|) + P_{i\rightarrow|f|}(-|k_{2y}|)}{\left|\frac{\partial(E_f - E_i)}{\partial k_{2y}}\right|_{k_{2y}}} \end{aligned} \tag{58}$$

For each k_{2x}, conservation of energy determines a corresponding value of $|k_{2y}|$. The condition that a real solution for $|k_{2y}|$ exists sets boundaries: a maximum and a minimum value for k_{2x}.

4.1.1 Scattering rate of $1TA \rightarrow 2ZA$

First we calculate the scattering rate of one transversal acoustic phonon to two flexural acoustic phonons. The only term of the interaction Hamiltonian contributing to the transition probability is 56c, since that is the only term for which $< f|H_{interaction}|i >$ has an equal amount of every type of phonon creation and annihilation operators. This probability can then be expressed as:

$$P_{1TA\rightarrow 2ZA} = |\frac{\mu}{2\sqrt{2A}} \frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k_3)\omega_0(k_2)\omega_T(k_1)}} t^T_\beta(k_1) k_{1\alpha}(k_{2\alpha}k_{3\beta} + k_{3\alpha}k_{2\beta})|^2 \tag{59}$$

This transition probability can be substituted in Fermi's Golden Rule (eq. 30) to obtain the scattering rate.
For integration as explained in 58, we first determine, at fixed k_{2x}, the value of k_{2y} satisfying the energy conservation and determine the derivative of $E_f - E_i$ at this value. This results in $k_{2y} = \pm\sqrt{-(k_{2x}^2 + \frac{k_1^2}{2} - k_1 k_{2x}) + \sqrt{\frac{\mu}{4\kappa}}k_1}$. This sets the boundaries for the integration over k_{2x} since all wavenumbers should remain real. From this, we can see that $k_{2x} \propto \sqrt{k_1}$. Since we look at the limit of $k_1 \rightarrow 0$, the leading terms are the ones of the lowest order in k_1 such that we neglect the higher order terms since they go faster to zero compared to the lowest order in k_1. This then gives:

$$k_{2y} = \pm\sqrt{-k_{2x}^2 + \sqrt{\frac{\mu}{4\kappa}}k_1} \tag{60}$$

and the following domain for k_{2x}:

$$k_{2x} = \left[-\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}, \sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}\right]. \tag{61}$$

This has the form of a general equation of a circle, as displayed in fig. 4, such that we can conclude that the possible decay directions can be displayed as a circle in the $\hat{x}\hat{y}$-plane with a radius of $\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}$ for $\vec{k_2}$ and where $\vec{k_3}$ is in opposite direction in the limit of $k_1 \rightarrow 0$. Here the final momenta $\vec{k_2}$ and $\vec{k_3}$ are bigger than the initial momentum $\vec{k_1}$.

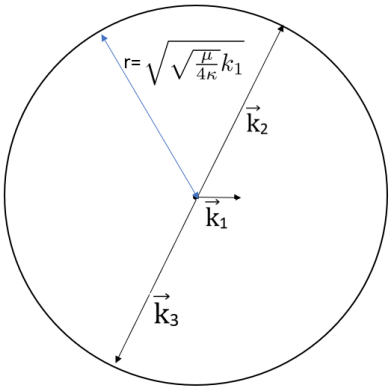

Figure 4: The phase space of the vectors $\vec{k_1}$, $\vec{k_2}$ and $\vec{k_3}$ for the process $1TA \rightarrow 2ZA$ in which for $\vec{k_1} \rightarrow 0$ the phase space is approximately a circle of radius $\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}$ where $\vec{k_2}$=- $\vec{k_3}$

The derivative of $E_f - E_i$ to k_{2y} at the value of k_{2y} (eq. 60) results in:

$$|\frac{\partial(E_f - E_i)}{\partial k_{2y}}|_{k_{2y}} = 4\hbar\sqrt{\frac{\kappa}{\rho}(-k_{2x}^2 + \sqrt{\frac{\mu}{4\kappa}}k_1)} \tag{62}$$

Since phonons are indistinguishable particles, we further normalize the scattering rate with a factor $\frac{1}{2}$.

Now we have determined all terms such that we can set up the integral as in eq. 58 and the decay rate can be rewritten as: :

$$\begin{aligned} R &= \frac{1}{2}\frac{2\pi}{\hbar}\sum_f P_{1TA\rightarrow 2ZA}\ \delta(E_f - E_i) \\ &= \frac{1}{2}\frac{2\pi}{\hbar}\int_{-\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}}^{\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}} \frac{\partial k_{2x}}{(2\pi)^2}\ \frac{P_{1TA\rightarrow 2ZA}(+k_{2y}) + P_{1TA\rightarrow 2ZA}(-k_{2y})}{4\hbar\sqrt{\frac{\kappa}{\rho}(-k_{2x}^2 + \sqrt{\frac{\mu}{4\kappa}}k_1)}} \end{aligned} \tag{63}$$

in which $P_{1TA\rightarrow 2ZA}$ is given in eq. 59. This results then in a scattering rate for one transversal acoustic phonon to two flexural phonons of:

$$R_{1TA\rightarrow 2ZA} = \frac{\hbar\mu^{\frac{3}{2}}k_1}{128\rho\kappa^{\frac{3}{2}}}. \tag{64}$$

The details of the derivation can be seen in Appendix 6.2. The scattering rate of this process increases when the initial momentum of the TA phonon increases, since it is proportional to k_1. This is due to the transition amplitude being proportional to $\sqrt{k_1}$ and not due to the density of final states (DOS) increasing when the initial momentum k_1 increases, since for a quadratic dispersion in two dimensions, the number of states between energy E and E+ dE is independent of E and thus

does not depend on k_1.
For the TA phonon to be well defined, the lifetime τ should be higher then the time needed to travel one wavelength, T, as was explained in eq. 32. For now we will assume that this is the only possible scattering decay for the TA phonon. The transversal acoustic phonon lifetime, which is equal to the inverse of the scattering rate, turns out to have a value of $\frac{3.8\times10^{-2}}{k_1}$. The lifetime should be longer than the time needed to travel a distance equal to one wavelength for the phonon to be well defined, therefore eq. 32 should be satisfied. The value of $\frac{\omega_T(k_1)}{R_{TA\rightarrow 2ZA}}$ of this process is equal to 5.3×10^2, which is bigger than 2π such that I conclude that the transversal phonon is well defined. However more scattering decays are possible, already when considering only the first order interactions, for the transversal acoustic phonon, which would decrease the true lifetime of the TA phonon. An example of another interesting decay of the TA phonon is the damping into two in-plane phonons, for which the linear spectrum imply that the momenta of final phonons are collinear to the initial momentum k_1.

4.1.2 Scattering rate of $1LA \rightarrow 2ZA$

The method for calculating the scattering rate of the process $1LA \rightarrow 2ZA$ is approximately the same. The terms of the interaction Hamiltonian (eq. 56) contributing to the transition probability are the terms 56b and 56c. The transition probability can be expressed as:

$$P_{1LA\rightarrow 2ZA} = \left| \frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k_3)\omega_0(k_2)\omega_L(k_1)}} \left(\frac{\mu}{2\sqrt{2A}} t^L_\beta(k_1) k_{1\alpha}(k_{2\alpha}k_{3\beta} + k_{3\alpha}k_{2\beta}) + \frac{\lambda}{2\sqrt{2A}}|k_1|(k_2\cdot k_3) \right) \right|^2 \tag{65}$$

which then can be substituted in Fermi's Golden Rule (eq. 30) to obtain the scattering rate of one longitudinal phonon to two acoustic flexural phonons. Here conservation of energy results in $k_{2y} = \pm\sqrt{-(k_{2x}^2 + \frac{k_1^2}{2} - k_1k_{2x}) + \sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}$, which also for this process sets the integration boundaries for k_{2x} and gives that $k_{2x} \propto \sqrt{k_1}$. Only considering the terms in the lowest order of k_1, since we are interested in the limit of $k_1 \rightarrow 0$, leads to the following:

$$k_{2y} = \pm\sqrt{-k_{2x}^2 + \sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1} \tag{66}$$

and the domain for k_{2x}:

$$k_{2x} = \left[-\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}, \sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1} \right] \tag{67}$$

Also for this decay we can conclude that the decay directions are all directions in the $\hat{x}\hat{y}$-plane, such that for it can be displayed as a circle with radius of $\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}$ (fig. 5). The direction of $\vec{k_3}$ is opposite to $\vec{k_2}$ with $k_{3x} = -k_{2x}$ and $k_{3y} = -k_{2y}$ in the limit of $k_1 \rightarrow 0$. The final momenta $\vec{k_2}$ and $\vec{k_3}$ are bigger than the initial momentum $\vec{k_1}$.

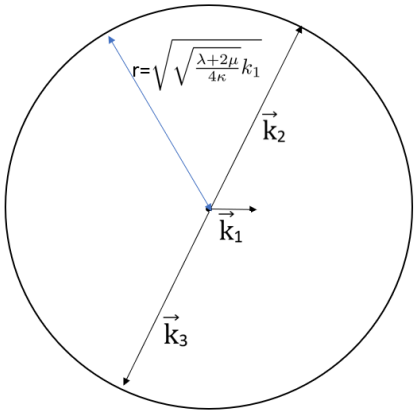

Figure 5: The phase space of the vectors $\vec{k_1}$, $\vec{k_2}$ and $\vec{k_3}$ for the process $1LA \rightarrow 2ZA$ in which for $\vec{k_1} \rightarrow 0$ the phase space is approximately a circle of radius $\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}$ where $\vec{k_2}$=- $\vec{k_3}$

Furthermore the derivative of $E_f - E_i$ to k_{2y} at the value of k_{2y} of eq. 66 is determined:

$$\left|\frac{\partial(E_f - E_i)}{\partial k_{2y}}\right|_{k_{2y}} = 4\hbar\sqrt{\frac{\kappa}{\rho}(\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1 - k_{2x}^2)} \tag{68}$$

Furthermore since phonons are bosonic, and are indistinguishable, we need to normalise the decay rate with a factor $\frac{1}{2}$. Now we have determined all terms needed to determine the scattering rate for the process of $1LA \rightarrow 2ZA$, which is:

$$\begin{aligned} R_{LA\rightarrow 2ZA} &= \frac{1}{2}\frac{2\pi}{\hbar}\sum_f P_{1LA\rightarrow 2ZA}\ \delta(E_f - E_i) \\ &= \frac{1}{2}\frac{2\pi}{\hbar}\int_{-\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}}^{\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}} \frac{\partial k_{2x}}{(2\pi)^2}\ \frac{P_{1LA\rightarrow 2ZA}(+k_{2y}) + P_{1LA\rightarrow 2ZA}(-k_{2y})}{4\hbar\sqrt{\frac{\kappa}{\rho}(-k_{2x}^2 + \sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1)}} \end{aligned} \tag{69}$$

This results in a scattering rate for longitudinal acoustic phonons to two flexural phonons of:

$$R_{1LA\rightarrow 2ZA} = \frac{\hbar k_1}{128\rho\kappa^{\frac{3}{2}}(\lambda+2\mu)^{\frac{1}{2}}}(3\mu^2 + 4\lambda\mu + 2\lambda^2) \tag{70}$$

The details of the derivation can be seen in Appendix 6.3.

For the LA phonon to be well defined, the lifetime τ should be higher then the time needed to travel one wavelength, T, as was explained in eq. 32. For now we will assume that $1LA \rightarrow 2ZA$ is the only possible scattering decay for the LA phonon, giving the it a lifetime of $\tau = \frac{1.2\times 10^{-2}}{k_1}$. From this we can conclude that the lifetime of the longitudinal acoustic phonon in this scattering process becomes smaller when the initial momentum is increased. Again the growth of the scattering rate can, instead, be attributed to the matrix element determining the transition amplitude, since it is

proportional to $\sqrt{k_1}$ and not due to the number of states increasing, since for particles having a quadratic dispersion in 2D, the density of final states turns out to be energy independent.
Furthermore the value of $\frac{\omega_L(k_1)}{R_{LA\rightarrow 2ZA}}$ of this process turns out to be approximately 256 which is higher than 2π and thus satisfies condition 32. Therefore I conclude that the longitudinal acoustic phonon is well defined in the assumption that this is the only process in which the LA phonon can decay in. But actually more scattering decays are possible, already when considering only the first order interactions, for the LA phonon. Therefore the true lifetime of the LA phonon will be lower.

4.1.3 Comparison between $1TA \rightarrow 2ZA$ and $1LA \rightarrow 2ZA$

As calculated, the phonon lifetime of the longitudinal acoustic phonon is lower than the lifetime of the transversal acoustic phonon when decaying in two flexural acoustic phonons. This can be explained by the fact that the interaction Hamiltonian (eq. 56) has more terms considering the scattering of longitudinal phonons than of transversal phonons, which increases the transition amplitude for the LA phonons.

4.2 Second Order Scattering Process: $1ZA \rightarrow 3ZA$

An example of an higher order scattering rate is the scattering of one flexural acoustic phonon to three out of plane phonons. We will obtain the scattering amplitude up to the second order interaction terms within a 'tree order' approximation. For this decay the initial state is defined as $|i>=\hat{a}^{\dagger}(k_1)|0>$ and the final state as $<f|=<0|\hat{a}(k_2)\hat{a}(k_3)\hat{a}(k_4)$ in the occupation number representation. Within this approximation, the decay can occur either directly or via a second-order process, in which an intermediate state $|m>$ is formed. The transition amplitude can be obtained by selecting tree-level terms from the second order perturbation formula, eq. 31.

The first order interaction, whose transition amplitude $<f|H_{int}|i>$ can be obtained by applying the interaction Hamiltonian once to the initial state, is determined by the term 56a of the interaction Hamiltonian. Which leads to:

$$\begin{aligned} <f|H_{int}|i> = \frac{\lambda+2\mu}{4A}\frac{\hbar^2}{\rho^2\sqrt{\omega_0(k_1)\omega_0(k_2)\omega_0(k_3)\omega_0(k_4)}} \\ \Big((k_{1\alpha}k_{2\alpha}k_{3\beta}k_{4\beta})+(k_{1\alpha}k_{3\alpha}k_{2\beta}k_{4\beta})+(k_{1\alpha}k_{4\alpha}k_{2\beta}k_{3\beta})\Big) \end{aligned} \tag{71}$$

By applying the interaction Hamiltonian twice we see that this decay process in second order can happen in two decay channels, when neglecting, within the tree-level approximation, intermediate states involving more then one virtual particle. The term 56c gives a nonzero scattering amplitude when applied twice, giving in the end our final state with in between an intermediate state $|p>=\hat{a}^{\dagger}(k_m)\hat{b}^{\dagger}(k_n)|0>$. The other possible decay channel results from the terms 56b and 56c of the interaction Hamiltonian with an intermediate state of $|q>=\hat{a}^{\dagger}(k_m)\hat{c}^{\dagger}(k_n)|0>$.

Then the second order transition amplitudes can be calculated according the following term in eq. 31, where is summed over the possible intermediate states:

$$\sum_m \frac{<f|H_{int}|m><m|H_{int}|i>}{E_i-E_m} \tag{72}$$

in which for the process with the intermediate state $|p>=\hat{a}^{\dagger}(k_m)\hat{b}^{\dagger}(k_n)|0>$:

$$<p|H_{int}|i>=\frac{\mu}{2\sqrt{2A}}\frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_T(k_n)\omega_0(k_m)\omega_0(k_1)}}\Big(k_{1\alpha}k_{n\alpha}k_{m\beta}t^T_{\beta}(k_n)+k_{n\alpha}k_{m\alpha}k_{1\beta}t^T_{\beta}(k_n)\Big)$$

and

$$\begin{aligned} <f|H_{int}|p> = \frac{\mu}{2\sqrt{2A}}\frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_T(k_n)}} \\ k_{n\alpha}t^T_{\beta}(k_n)\Big(\delta_{k_4,k_m}\frac{(k_{2\alpha}k_{3\beta}+k_{3\alpha}k_{2\beta})}{\sqrt{\omega_0(k_2)\omega_0(k3)}})+\delta_{k_2,k_m}\frac{(k_{4\alpha}k_{3\beta}+k_{3\alpha}k_{4\beta})}{\sqrt{\omega_0(\omega_0(k_3)\omega_0(k_4)}}+\delta_{k_3,k_m}\frac{(k_{2\alpha}k_{4\beta}+k_{4\alpha}k_{2\beta})}{\sqrt{\omega_0(k_2)\omega_0(k_4)}}\Big) \end{aligned}$$

These two transition amplitudes are normalized by the difference between the energy of the intermediate state and the initial state or the final state, which are equal in energy.

$$\frac{1}{E_i-E_p}=\frac{1}{\hbar\Big(\sqrt{\frac{\kappa k_1^4}{\rho}}-\sqrt{\frac{\mu k_n^2}{\rho}}-\sqrt{\frac{\kappa k_m^4}{\rho}}\Big)}$$

And for the intermediate state $|q>=\hat{a}^{\dagger}(k_m):\hat{c}^{\dagger}(k_n)|0>$

$$< q|H_{int}|i> = \frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k_1)\omega_0(k_m)\omega_L(k_n)}}\Bigg(\frac{\lambda}{2\sqrt{2A}}|k_n|(k_1\cdot k_m)+ \frac{\mu}{2\sqrt{2A}}\Big(t^L_{\alpha}(k_n)k_{m\alpha}k_{1\beta}k_{n\beta}+t^L_{\beta}(k_n)k_{1\beta}k_{m\alpha}k_{n\alpha}\Big)\Bigg)$$

and

$$\begin{aligned}< f|H_{int}|q> = {}& \frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_L(k_n)}} \\ & \Big(\frac{\mu}{2\sqrt{2A}}t^L_{\beta}(k_n)k_{n\alpha}\big(\delta_{k_4,k_m}\frac{(k_{2\alpha}k_{3\beta}+k_{3\alpha}k_{2\beta})}{\sqrt{\omega_0(k_2)\omega_0(k_3)}}+\delta_{k_2,k_m}\frac{(k_{4\alpha}k_{3\beta}+k_{3\alpha}k_{4\beta})}{\sqrt{\omega_0(k_3)\omega_0(k_4)}}+\delta_{k_3,k_m}\frac{(k_{2\alpha}k_{4\beta}+k_{4\alpha}k_{2\beta})\big)}{\sqrt{\omega_0(k_2)\omega(k_4)}} \\ & +\frac{\lambda}{2\sqrt{2A}}t^L_{\alpha}(k_n)k_{n\alpha}\big(\delta_{k_4,k_m}\frac{k_{2\beta}k_{3\beta}}{\sqrt{\omega_0(k_2)\omega_0(k_3)}}+\delta_{k_3,k_m}\frac{k_{2\beta}k_{4\beta}}{\omega_0(k_2)\omega_0(k_4)}+\delta_{k_2,k_m}\frac{k_{4\beta}k_{3\beta}}{\omega_0(k_3)\omega_0(k_4)}\big)\Big)\end{aligned}$$

And here also these transition amplitudes are normalized by the difference of the initial state and the intermediate state.

$$\frac{1}{E_i-E_q}=\frac{1}{\hbar\Big(\sqrt{\frac{\kappa k_1^4}{\rho}}-\sqrt{\frac{\lambda+2\mu k_n^2}{\rho}}-\sqrt{\frac{\kappa k_m^4}{\rho}}\Big)}$$

The total transition amplitude of the decay of one ZA phonon to three ZA phonons is given by the sum of the first order transition amplitude and the two second order transition amplitudes (see eq. 31), such that the scattering rate of this decay can be calculated by Fermi's Golden Rule(eq. 30) :

$$R_{ZA\rightarrow 3ZA}=\frac{2\pi}{\hbar}\sum_f\delta(E_f-E_i)\left|< f|H_{int}|i> + \frac{< f|H_{int}|q>< q|H_{int}|i>}{E_i-E_q}+\frac{< f|H_{int}|p>< p|H_{int}|i>}{E_i-E_p}\right|^2 \tag{73}$$

in which with the conservation of momentum the following can be used to reduce the number of variables:

$$\begin{aligned}\vec{k_1} &= \vec{k_2}+\vec{k_3}+\vec{k_4} \\ &= \vec{k_m}+\vec{k_n}\end{aligned}$$

$$\vec{k_n}=\frac{1}{3}\Big(\delta_{k_m,k_4}\big(\vec{k_2}+\vec{k_3}\big)+\delta_{k_m,k_3}\big(\vec{k_2}+\vec{k_4}\big)+\delta_{k_m,k_2}\big(\vec{k_4}+\vec{k_3}\big)\Big)$$

Due to the limited time I have for this bachelor project, I could not finish this calculation.

5 Conclusion

The interaction Hamiltonian within the continuum elasticity theory is derived and expressed in phonon creation and annihilation operators (eq. 56)
We determined the scattering rates of $1TA \to 2ZA$ and $1LA \to 2ZA$ with Fermi's Golden Rule, which we did by only considering the leading order, which is the lowest order, in k_1 in the limit of $k_1 \to 0$. From this I could conclude that for both decay processes $1LA \to 2ZA$ and $1TA \to 2ZA$ the decay rate is proportional to the initial momentum k_1, which came from the transition probability being proportional to k_1. Since the interaction Hamiltonian consists of more terms describing the scattering of longitudinal phonons than of transversal phonons, the scattering rate of LA phonons is higher than of TA phonons.
Lastly we concluded that both the TA and LA phonon are well defined at 0K, when considering only the decay in two ZA phonons. However the true lifetime of the LA and TA phonon are lower since more scattering processes are possible then only the decay in two flexural phonons.

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6 Appendix

6.1 Mathematical identities

$$\int_{-\infty}^{\infty} dx\ \frac{\sin^2(ax)}{x^2} = a\pi \tag{74}$$

$$\sum_f = A \int \frac{d^2\vec{k_2}}{(2\pi)^2} \tag{75}$$

$$\int dx\ \delta(g(x))\ f(x) = \sum_{x_i} \frac{f(x_i)}{|\frac{\partial g(x)}{\partial x}|_{xi}} \text{ where } x_i \text{ is such that } g(x_i) = 0 \text{ following from } \delta(g(x)). \tag{76}$$

6.2 Derivation of the scattering rate of $1TA \to 2ZA$

$$R_{1TA\to 2ZA} = \frac{2\pi}{\hbar} \sum_f \delta(E_f - E_i) \left| \frac{\mu}{2\sqrt{2A}} \frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}} \sqrt{\omega_0(k_2)\omega_0(k_3)\omega_T(k_1)}} \left((t^T(k_1)\cdot k_3)(k_1\cdot k_2) + (t^T(k_1)\cdot k_2)(k_1\cdot k_3) \right) \right|^2$$

$$= \frac{2\pi\mu^2\hbar^2}{8A\omega_T(k_1)\rho^3} \sum_f \delta(E_f - E_i) \frac{(k_1^2 k_{2x}^2 k_{3y}^2 + k_1^2 k_{3x}^2 k_{2y}^2 + 2k_1^2 k_{2x} k_{2y} k_{3x} k_{3y})}{\frac{\kappa}{\rho}(k_{2x}^2 + k_{2y}^2)(k_{3x}^2 + k_{3y}^2)}$$

For going to the next step, the degrees of freedom is lowered by considering momentum conservation with which we found $k_{3y} = -k_{2y}$ and $k_{3x} = k_1 - k_{2x}$.

$$= \frac{2\pi\mu^2\hbar^2}{8A\omega_T(k_1)\rho^3} \sum_f \delta(E_f - E_i) \frac{(3k_1^2 k_{2x}^2 k_{2y}^2 + k_1^2 k_{2y}^2 (k_1^2 + k_{2x}^2 - 2k_1 k_{2x})}{(k_{2x}^2 + k_{2y}^2)^2}$$

Then only the lowest order of k_1 is considered, for which first we had to determine that $k_{2x} \propto \sqrt{k_1}$ which could be done by considering the conservation of energy.

$$= \frac{\pi\hbar^2\mu^2}{\rho^2\kappa A\omega_T(k_1)} \sum_f \delta(E_f - E_i) \frac{k_1^2 k_{2x}^2 k_{2y}^2}{(k_{2x}^2 + k_{2y}^2)^2}$$

The sum over final states is replaced by $A\int \frac{d^2\vec{k_2}}{(2\pi)^2}$. Furthermore the integral over k_{2y} is done as explained in eq. 58. $\delta(E_f - E_i)$ gives $\hbar\left(\sqrt{\frac{\kappa}{\rho}(k_{2x}^2 + k_{2y}^2)^2} + \sqrt{\frac{\kappa}{\rho}(k_{3x}^2 + k_{3y}^2)^2} - \sqrt{\frac{\mu}{\rho}}k_1\right) = 0$. This leads to the expression for k_{2y} in eq. 60 and the boundaries for k_{2x} in eq. 61.

$$= \frac{\hbar\mu}{2\pi\rho^{\frac{3}{2}}\kappa^{\frac{1}{2}}k_1^2\omega_T(k_1)} \int_{\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}}^{\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1}} dk_{2x} \frac{\sqrt{\frac{\mu}{4\kappa}}k_1^3 k_{2x}^2 - k_1^2 k_{2x}^4}{\sqrt{\sqrt{\frac{\mu}{4\kappa}}k_1 - k_{2x}^2}}$$

By substitution of $x = \sqrt{a}\sin(u)$ in which a is given by $\sqrt{\frac{\mu}{4\kappa}}k_1$ the integral can be calculated, leading to

$$= \frac{\hbar\mu}{2\pi\rho^{\frac{3}{2}}\kappa^{\frac{1}{2}}k_1^2\omega_T(k_1)}\left(\frac{\mu}{4\kappa}k_1^4\sin(1)^{-1} - \frac{3}{4}\frac{\mu}{4\kappa}k_1^4\sin(1)^{-1}\right)$$
$$= \frac{\hbar\mu^{\frac{3}{2}}k_1}{64\rho\kappa^{\frac{3}{2}}}$$

Since phonons are bosonic and thus indistinguishable particles, the scattering rate should be normalized with a factor $\frac{1}{2}$.

$$= \frac{\hbar\mu^{\frac{3}{2}}k_1}{128\rho\kappa^{\frac{3}{2}}}$$

6.3 Derivation of the scattering rate of $1LA \to 2ZA$

$$R_{1LA\to 2ZA} = \frac{2\pi}{\hbar}\sum_f \delta(E_f - E_i)\left|\frac{\hbar^{\frac{3}{2}}}{\rho^{\frac{3}{2}}\sqrt{\omega_0(k_3)\omega_0(k_2)\omega_L(k_1)}}\left(\frac{\mu}{2\sqrt{2A}}\Big((t^L(k1)\cdot k_2)(k_1\cdot k_3) + (t^L(k1)\cdot k_3)(k_1\cdot k_2)\Big)\right.\right.$$
$$\left.\left. + \frac{\lambda}{2\sqrt{2A}}|k_1|(k_2\cdot k_3)\Big)\right|^2$$
$$= \frac{2\pi}{\hbar}\frac{\hbar^3}{\rho^3\omega_L(k_1)}\sum_f \delta(E_f - E_i)\frac{1}{\frac{\kappa}{\rho}(k_{2x}^2+k_{2y}^2)(k_{3x}^2+k_{3y}^2)}\Big(\frac{\mu^2}{8A}4k_{2x}^2k_{3x}^2k_1^2 + \frac{\lambda^2}{8A}k_1^2(k_{2x}k_{3x}+k_{2y}k_{3y})^2$$
$$+ \frac{\lambda\mu}{2A}(k_{2x}k_1k_{3x})(|k_1|(k_{2x}k_{3x}+k_{2y}k_{3y})\Big)$$

With momentum conservation we can rewrite $k_{3x} = k_1 - k_{2x}$ and $k_{3y} = -k_{2y}$. By considering only the lowest order in k_1, we get

$$= \frac{\pi\hbar^2}{4A\rho^2\kappa\omega_L(k_1)}\sum_f \delta(E_f - E_i)\frac{(4\mu^2+\lambda^2+4\lambda\mu)k_1^2k_{2x}^4 + (2\lambda^2+4\lambda\mu)k_1^2k_{2x}^2k_{2y}^2 + \lambda^2k_1^2k_{2y}^4}{k_{2x}^4+k_{2y}^4+2k_{2x}^2k_{2y}^2}$$

The sum over final states is replaced by $A\int\frac{d^2\vec{k_2}}{(2\pi)^2}$. Furthermore the integral over k_{2y} is done as explained in eq. 58, where energy conservation gives $\hbar\left(\sqrt{\frac{\kappa}{\rho}(k_{2x}^2+k_{2y}^2)^2} + \sqrt{\frac{\kappa}{\rho}(k_{3x}^2+k_{3y}^2)^2} - \sqrt{\frac{\lambda+2\mu}{\rho}}k_1\right) = 0$. This leads to the expression for $k_{2y} = \pm\sqrt{-k_{2x}^2 + \sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}$ as eq. 66 and the boundaries for k_{2x} given by $\Big[-\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}, \sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}\Big]$ as in eq. 67.

$$= \frac{\hbar}{32\pi\rho^{\frac{3}{2}}\kappa^{\frac{3}{2}}\omega_L(k_1)}\frac{4\kappa}{(\lambda+2\mu)k_1^2}\int_{-\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}}^{\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1}} dk_{2x}\frac{\Big(4\mu^4k_1^2k_{2x}^2 + 4\lambda\mu\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1^3k_{2x}^2 + \lambda^2\frac{\lambda+2\mu}{4\kappa}k_1^4\Big)}{\sqrt{\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1 - k_{2x}^2}}$$

By substitution of $x = \sqrt{a}\sin(u)$ in which a is given by $\sqrt{\frac{\lambda+2\mu}{4\kappa}}k_1$ in the integral can be calculated, leading to

$$= \frac{\hbar}{8\pi\rho^{\frac{3}{2}}\kappa^{\frac{1}{2}}\omega_L(k_1)(\lambda+2\mu)k_1^2}\Big(3\mu^2k_1^4\frac{(\lambda+2\mu)}{4\kappa}\sin^{-1}(1) + 4\lambda\mu\frac{(\lambda+2\mu)}{4\kappa}k_1^4\sin^{-1}(1) + 2\lambda^2\frac{(\lambda+2\mu)}{4\kappa}k_1^4\sin^{-}1(1)\Big)$$

Since phonons are bosonic, they are indistinguishable and therefore the scattering rate should be normalized with a factor $\frac{1}{2}$.

$$= \frac{\hbar k_1}{128\rho\kappa^{\frac{3}{2}}(\lambda+2\mu)^{\frac{1}{2}}}(3\mu^2 + 4\lambda\mu + 2\lambda^2)$$