# INVESTIGATION OF THE THERMAL TRANSPORT IN SUPERCONDUCTING NIOBIUM AND TANTALUM

By

Peng Xu

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## ABSTRACT

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Superconducting radio frequency (SRF) cavities fabricated from bulk high purity niobium (Nb) are increasingly used for particle accelerators to achieve continuous operation. Even in the superconducting regime, residual resistance and small imperfections on the RF surface can dissipate energy and cause local heating that leads to cavity quench. Large values of thermal conductivity can mitigate local temperature excursions and prevent cavity quench, thus improving cavity performance. Understanding thermal transport in bulk and thin film superconducting Nb may guide thermal design of current and next generation SRF cavities.

The thermal conductivity of metals is composed of electronic and lattice (phonon) components. In normal conductors, the electronic component dominates, and in superconducting metals, as the temperature drops below the critical temperature, phonons become increasingly important carriers of thermal energy. A widely used model of thermal conductivity in superconductors omits explicit accounting of the effect of dislocations, which result from deformation. Here, this model is extended by accounting for the effects of phonons scattered by dislocations independent from boundary scattering. This extended model agrees better with measurements of thermal conductivity in deformed Nb samples, especially at temperatures *T* less than 3 K. An apparent threshold of dislocation density  $N_d$  is found to be  $N_d = O(10^{12})$  m<sup>-2</sup> for Nb and when applied to tantalum (Ta), it is  $N_d = O(10^{11})$  m<sup>-2</sup>. There is little contribution to the thermal conductivity when the dislocation density is less than this threshold. This model can also be used to estimate the dislocation density by fitting measured values of thermal conductivity.

Examination of thermal conductivity data for superconducting Nb shows that there is often a local maximum, a so-called phonon peak,  $k_{pp}$ . The temperature at which this  $k_{pp}$  occurs  $T_{pp}$  is between 1.72 K and 2.35 K and shifts for samples after deformation. It is well known that the

magnitude of  $k_{pp}$  decreases as the material is deformed, and hence with increasing  $N_d$ . Less cited is that  $T_{pp}$  increases with increasing  $N_d$ . This may affect the operating temperature of an SRF cavity. At a certain level deformation (i.e., 4.7% deformation for a residual resistivity ratio RRR=185), the phonon peak disappears. More deformation is needed for higher RRR, (i.e., greater purity).

The models discussed above require estimating several parameters from thermal conductivity measurements and may be best suited to explaining the relative importance of the several scattering mechanisms. For predicting thermal conductivity from basic material variables, the Boltzmann transport equation (BTE) is solved by two methods to predict the lattice component of thermal conductivity. One method uses a substitution of variables from frequency to wavevector in the Callaway model to include the nonlinear phonon dispersion relationship for the longitudinal acoustic (LA) and transverse acoustic (TA) phonon polarizations. This model incorporates a relaxation time approximation using Matthiessen's rule to consider phonon scattering by electrons, boundaries, and dislocations. Another method to predict the lattice thermal conductivity uses an energy-based, variance-reduced Monte Carlo (MC) solution to the BTE for phonons. The MC solution allows more general consideration of the individual scattering mechanisms. It may also be generalized for more complex geometries. The MC solution technique was first verified by comparing the predicted thermal conductivity in bulk Si and Si nanowires with experimental results. Both solutions of the BTE for the lattice thermal conductivity of undeformed and deformed superconducting Nb agreed well with experimental values. The MC model was also used to demonstrate that interstitial impurities must be near saturation to change the lattice thermal conductivity of Nb. The MC solution was also effective in predicting the lattice thermal conductivity of superconducting Ta, with the appropriate change in dispersion relation and other material parameters.

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# TABLE OF CONTENTS

LIST OF	F TABLES	• viii
LIST OF	F FIGURES	• ix
CHAPT	ER 1 INTRODUCTION	• 1
CHAPT 2.1 2.2 2.3	ER 2       REVIEW OF THERMAL CONDUCTIVITY OF NB	. 4 . 10 . 23 . 31
CHAPT	ER 3 EFFECT OF DEFORMATION ON THE THERMAL CONDUCTIVITY	
3.1 3.2	AND PHONON PEAK TEMPERATURE OF NB         Introduction         Analysis         3.2.1         Scaled sensitivity coefficient         3.2.1.1         Scaled sensitivity coefficient of boundary scattering         3.2.1.2         Scaled sensitivity coefficient of normal electron scattering         3.2.1.3	<ul> <li>32</li> <li>32</li> <li>32</li> <li>34</li> <li>35</li> <li>35</li> <li>35</li> <li>36</li> </ul>
3.3	Results and Discussion	. 38
3.4	Conclusion	. 43
CHAPT	ER 4 MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC-	48
CHAPT 4.1	ER 4    MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB      Introduction    Introduction	• 48 • 48
CHAPT 4.1 4.2	ER 4    MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB      Introduction    Introduction      Boltzmann Transport Equation	• 48 • 48 • 49
CHAPT 4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction       Introduction         Boltzmann Transport Equation       Introduction         Monte Carlo Solution Technique       Introduction	<ul> <li>48</li> <li>48</li> <li>48</li> <li>49</li> <li>50</li> </ul>
4.1 4.2 4.3	ER 4    MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB      Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> </ul>
CHAPT 4.1 4.2 4.3	ER 4    MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB      Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> </ul>
4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> </ul>
4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> </ul>
4.1 4.2 4.3	ER 4MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NBIntroduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> </ul>
4.1 4.2 4.3	ER 4MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NBIntroduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> <li>54</li> </ul>
4.1 4.2 4.3	ER 4MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NBIntroduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> <li>54</li> </ul>
4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> <li>54</li> <li>54</li> <li>55</li> </ul>
4.1 4.2 4.3	ER 4MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NBIntroduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>54</li> <li>55</li> <li>55</li> </ul>
4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> <li>54</li> <li>55</li> <li>55</li> <li>56</li> </ul>
4.1 4.2 4.3	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>54</li> <li>55</li> <li>55</li> <li>56</li> <li>57</li> </ul>
4.1 4.2 4.3 4.4	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>53</li> <li>54</li> <li>55</li> <li>56</li> <li>57</li> <li>57</li> </ul>
4.1 4.2 4.3 4.4	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>54</li> <li>55</li> <li>56</li> <li>57</li> <li>57</li> <li>57</li> </ul>
4.1 4.2 4.3 4.4	ER 4       MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUC- TIVITY OF NB         Introduction	<ul> <li>48</li> <li>48</li> <li>49</li> <li>50</li> <li>50</li> <li>51</li> <li>52</li> <li>53</li> <li>54</li> <li>55</li> <li>56</li> <li>57</li> <li>57</li> <li>57</li> <li>58</li> </ul>

	4.4.4 Size effect on thermal conductivity of Si nanowires	60
	4.4.5 Simulation of the thermal conductivity of Nb	61
	4.4.5.1 Simulation of the lattice thermal conductivity of undeformed Nb .	62
	4.4.5.2 Simulation of the lattice thermal conductivity of deformed Nb	64
4.5	Conclusion	66
СНАРТ	ER 5 EFFECT OF IMPURITIES ON THE LATTICE THERMAL CONDUC-	
		68
5.1	Residual resistance ratio (RRR)	68
5.2	Effect of oxygen diffusion	71
5.3	Effect of hydrogen diffusion	72
5.4	Effect of nitrogen diffusion	73
55	Substitutional atoms of different mass	73
5.5	Point defects in the MC Model	74
5.0 5.7	Monte Carlo simulation considering impurity scattering	74
5.8	Phonon-impurity scattering	78
5.0	Effect of Grain Boundary scattering on the lattice thermal conductivity of super	70
5.9	conducting Nb	80
	5.0.1 Modele	00 00
5 10	S.9.1 Models	00 00
5.10		02
CHAPT	ER 6 CALCULATION OF THE LATTICE THERMAL CONDUCTIVITY OF	
	NB CONSIDERING PHONON DISPERSION	84
6.1	Introduction	84
6.2	Model	84
0.2	6.2.1 Phonon dispersion of Nb	86
	6.2.2 Phonon-boundary scattering	86
	6.2.3 Phonon-electron scattering	86
	6.2.4 Phonon-dislocation scattering	87
	6.2.5 Total relaxation time	88
63	Results and Discussions	88
6.5		01
0.4		91
CHAPT	ER 7 THERMAL CONDUCTIVITY OF SUPERCONDUCTING TANTALUM .	93
7.1	Introduction	93
7.2	Monte Carlo simulation	93
7.3	Comparison of total $k$ between Nb and Ta $\ldots$	98
	7.3.1 Comparison of simulated lattice $k$ between undeformed Nb and Ta	99
	7.3.2 Comparison of simulated lattice $k$ between deformed Nb and Ta	100
7.4	Conclusion	101
		102
CHAPL		103
APPEN	DIX	105
BIBLIO	GRAPHY	191

# LIST OF TABLES

Table 2.1:	Chronology of measurements of the thermal conductivity of superconducting Nb. RRR values marked with asterisks (*) are estimated using measured $k(4.2 \text{ K}) \times 4$	21
Table 2.2:	Numerical integration results for $I_1(z)$ and $I_2(z)$ [1]	29
Table 2.3:	Research progress on modeling $k$ of superconducting Nb	30
Table 4.1:	Parameters for the samples used in the simulation	61
Table 5.1:	Contributions of different impurities to $\frac{\partial \rho_i}{\partial C_i}$ [1]	69
Table 5.2:	Typical RRR values of Nb in relation to processings [2], where Cavities and Samples refer to post purified Nb origins, Other refers to other preparations	70
Table 7.1:	Parameters for the Ta samples used in the simulation [3], where the size refers to the diameter of the cylinder. All of the three Ta samples were deformed at temperature higher or equal to 295 K, therefore they are likely to have edge dislocations.	94

# LIST OF FIGURES

Figure 1.1:	A typical example of the thermal conductivity of single crystal supercon- ducting Nb sample from 0.3 K to 5 K with different levels of uniaxial strain, replotted from [4]. Undeformed state refers to Cauchy strain of 0%, and 1%, 2.4%, 4.7%, and 8.8% refer to the corresponding amounts of Cauchy strain. The thermal conductivity at temperature lower than 3 K decreases with the increase of strain. The phonon peak appears at 0%, 1%, and 2.4% deforma- tion and disappears after 4.7% deformation. The thermal conductivity at the phonon peak decreases the most after deformation compared with the thermal conductivity at other temperatures.	2
Figure 2.1:	An example of the sketch of a 9-cell TESLA accelerating structure [5]. The outside surface of the cavity is immersed into liquid He to keep Nb in superconducting temperature. Charged particles move inside the cavity to be accelerated by the magnetic filed.	4
Figure 2.2:	Picture of the delamination crack. The crack is located approximately in the middle of the Nb sheet, spiting the sheet into two parts.	5
Figure 2.3:	Microscopic image of the delamination crack. The crack propagates around 25 mm along the circumference, the thickness of the Nb sheet is around 2 mm, the crack splits the Nb sheet by half	5
Figure 2.4:	The contributions of the normal electron thermal conductivity $k_e$ and the lattice thermal conductivity $k_g$ to the overall thermal conductivity $k = k_e + k_g$ . At temperature lower than 2 K, $k_g$ dominates the total thermal conductivity and $k_e$ dominates the thermal conductivity for temperature higher than 3 K. For this Nb with relatively few dislocations, there is a significant phonon peak.	7
Figure 2.5:	Illustration of the roles of dislocation density and RRR on polycrystalline [6, 7] and single crystal Nb [4]. Thermal conductivity at temperature T=4.2 K is proportional to the RRR. However, at the temperatures close to 2 K, the strain has more effect on the thermal conductivity than the RRR. Of particular note is that a single sample is measured in its as-received state at RRR=390, after annealing at 1200 °C for two hours, but with a Ti getter, the purity is	
	reduced to RRR=68, but a phonon peak develops.	9

Figure 2.6:	A typical example of the thermal conductivity of single crystal supercon- ducting Nb sample with screw dislocations from 0.3 K to 5 K with different levels of uniaxial strain, replotted from [4]. Undeformed state refers to 0 Cauchy strain, and 3%, 7.3%, and 14.7% deformation correspond to the same amount of Cauchy strain. The thermal conductivity at temperature lower than 3 K decreases with the increase of strain. The phonon peak appears at 0% deformation and 3% at about 1.8 K and 2 K separately. There is nearly no phonon peak after 7.3% deformation and 14.7% deformation. The thermal conductivity data at temperatures higher than 3.5 K are the same for different amount of deformations	17
Figure 2.7:	Comparison between fitting with or without the dislocation term for deformed sample [4]. The estimated value of dislocation density is $N_d = 3.83 \times 10^{14} \text{ m}^{-2}$ .	28
Figure 3.1:	Measured phonon peak temperatures $T_{pp}$ for undeformed, deformed, and annealed specimens as a function of the ratio of the thermal conductivity at the phonon peak $k_{pp}$ to the local minimum thermal conductivity $k_{lm}$	33
Figure 3.2:	The scaled sensitivity coefficients of phonons scattered by boundary (parameter B) as a function of temperature. The B values vary from $2 \times 10^3 \text{ Wm}^{-2} \text{K}^{-4}$ to $10 \times 10^3 \text{ Wm}^{-2} \text{K}^{-4}$ .	36
Figure 3.3:	The scaled sensitivity coefficients of phonons scattered by normal electrons (parameter D) as a function of temperature. The D values vary from $2 \times 10^{-3}$ WK <sup>3</sup> m <sup>-1</sup> to $10 \times 10^{-3}$ WK <sup>3</sup> m <sup>-1</sup>	37
Figure 3.4:	The scaled sensitivity coefficients of phonons scattered by dislocations (parameter $N_d$ ) as a function of temperature. The $N_d$ values vary from $2 \times 10^{12}$ $m^{-2}$ to $10 \times 10^{12} m^{-2}$ .	38
Figure 3.5:	Comparison between fit with or without dislocation term for an undeformed sample from [4]. The estimated value of dislocation density is $N_d = 4.67 \times 10^{12} \text{ m}^{-2}$ . Both the fitted lines for temperatures lower than 1 K and higher than 2.5 K agree well with the experimental data. At 1.5 K < T < 2 K, fitting with the dislocation term has k values lower than the experimental results, while fitting without dislocation term has k values higher than the experimental results. The fit with or without the dislocation term provides similar accuracy for undeformed samples.	39

Figure 3.6:	Comparison between fitting with or without dislocation term for deformed sample [4]. The estimated value of dislocation density is $N_d = 3.83 \times 10^{14}$ m <sup>-2</sup> . Both the fitted lines for temperatures lower than 1 K and higher than 2.5 K agree well with the experimental data. At 1 K < T < 2.5 K, fitting with dislocation term matches well with the experimental results, while fitting without the dislocation term deviates from the experimental results. The fit with the dislocation term improves the accuracy for samples after deformations	40
Figure 3.7:	Differences between measurement data and the fit results of thermal conduc- tivity with or without the dislocation term for an undeformed sample [4]. Here the differences, which are also called the residuals, are calculated as $k_m - k_f$ , where $k_m$ is the measured thermal conductivity and $k_f$ is the fitted data.	40
Figure 3.8:	Differences between measurement data and the fit results of thermal con- ductivity with or without the dislocation term for a sample after 14.7% de- formation [4]. Here the differences, which are also called the residuals, are calculated as $k_m - k_f$ , where $k_m$ is the measured thermal conductivity and $k_f$ is the fitted data.	42
Figure 3.9:	Thermal conductivity of a sample from [4] (an example of predominantly screw dislocation) using curve fitting including the effect of dislocation. Here the dislocation density is estimated from Eq. 2.4 for the Cauchy strain $\epsilon = 0\%$ , 3%, 7.3%, 10.3%, and 14.7%, respectively. Wasserbäch [4] provided a mean value of the dislocation density to be $0.8 - 1 \times 10^{14} \text{ m}^{-2}$ , and the dislocation density measured at 14.7% is on the order of magnitude of $10^{14} \text{ m}^{-2}$	43
Figure 3.10:	Thermal conductivity of a sample from [4] (an example of predominantly edge dislocation) using curve fitting including the effect of dislocation. Here the dislocation density is estimated from Eq. 2.4 for the Cauchy strain $\epsilon = 0\%$ , 4%, and 8% respectively. Wasserbäch [4] provided a mean value of the dislocation density to be $1 - 2 \times 10^{14}$ m <sup>-2</sup> , and the dislocation density measured at 8% is on the order of magnitude of $10^{14}$ m <sup>-2</sup> .	44
Figure 3.11:	The effect of dislocation density on thermal conductivity of Nb at temperature between 1.5 K and 4.5 K. Equation 2.4 is used to fit the experimental results from [8]. The results are obtained by using the fitted parameters ( <i>e.g.</i> , <i>B</i> , <i>D</i> , <i>a</i> ) and changing $N_d$ .	45

Figure 3.12:	Comparison between phonon boundary thermal resistance and phonon dislo-	
	cation resistance for different dislocation density by using the fitted parameters	
	$(e.g., B, D, a)$ and changing $N_d$	46

Figure 3.13:	The ratio of dislocation density after deformation to that before deformation as a function of uniaxial straining, estimated by fitting the thermal conductivity from experimental results [4]. Samples H1, K6, H2, and K10 are deformed at temperatures of 77 K, 195 K, 295 K, and 1470 K, respectively. H1 and K6 had predominantly screw dislocations, H2 and K10 had predominantly edge dislocation.	47
Figure 4.1:	Comparison of the temperature profile obtained using the Monte Carlo model with the usual Fourier conduction results in the diffusion limit. Six Monte Carlo simulations have been carried out to demonstrate the repeatability of the model, and the scatter in the results are shown	58
Figure 4.2:	Comparison of the temperature profile by Monte Carlo method with Stefen- Boltzmann equation in the ballistic limit. Six simulations have been carried out to demonstrate the repeatability of the Monte Carlo model	59
Figure 4.3:	Comparison of the thermal conductivity of bulk Si between different methods. The solid line is Eq. 4.22, the circles come from Monte Carlo simulation by Lacroix <i>et al.</i> [9], and the stars are the current simulation results	60
Figure 4.4:	Comparison of the thermal conductivity of Si nanowire between current simulations (solid lines) and the data (triangles) from Li <i>et al.</i> [10]. Six simulations have been carried out and the error bars were plotted for the Monte Carlo simulation results.	61
Figure 4.5:	Phonon dispersion relation of Nb in the [1 0 0] direction, replotted from reference [11], where <i>a</i> is the lattice parameter, which refers to the physical dimension of unit cell in a crystal lattice, for Nb, $a = 3.3 \times 10^{-10}$ m	62
Figure 4.6:	Simulation results of lattice $k$ of undeformed Nb or Nb after heat treatment. Data used from undeformed samples from reference [4] and the sample after heat treatment of reference [12]	63
Figure 4.7:	Simulation results of lattice thermal conductivity of Nb LS 1 from [4], an example of screw dislocations, for several strains.	65
Figure 4.8:	Simulation results of lattice thermal conductivity of Nb LS 2, from [4] an example of edge dislocations, for several strains.	66
Figure 5.1:	Typical thermal conductivity data [4, 7, 13] related with RRR and strain, thermal conductivity increases with RRR at 4.2 K, while at 2 K, there is no relation between RRR and thermal conductivity. The phonon peak usually exists for appropriately heat treated sample while strain may destroy the phonon peak. (This figure is a repeat of Fig. 2.5), included here for the reader's convenience.	70

Figure 5.2:	Temperature profile of Si for different levels of impurities from $10^{23}$ m <sup>-3</sup> to $10^{28}$ m <sup>-3</sup> . The temperature profile changes gradually from the ballistic limit for the lowest defect density $10^{23}$ m <sup>-3</sup> to diffusion limit at the highest defect density $10^{28}$ m <sup>-3</sup> .	 75
Figure 5.3:	Simulation of the lattice thermal conductivity of Nb with different levels of H impurities compared with fitting results without H impurities. The lattice thermal conductivity values are similar for low concentrations of H impurities ( <i>i.e.</i> No impurity, $10^{26}$ m <sup>-3</sup> , and $10^{28}$ m <sup>-3</sup> ), but at the impurity concentration of $10^{29}$ m <sup>-3</sup> , the lattice thermal conductivity decreased by 20% at the phonon peak.	 76
Figure 5.4:	Simulation of the lattice thermal conductivity of Nb with different levels of N impurities compared with fitting results without N impurities. The lattice thermal conductivity values are similar for low concentrations of N impurities ( <i>i.e.</i> No impurity, $10^{26}$ m <sup>-3</sup> , and $10^{27}$ m <sup>-3</sup> ), but at the impurity concentration of $10^{28}$ m <sup>-3</sup> , the lattice thermal conductivity decreased by 15% at the phonon peak.	 77
Figure 5.5:	Lattice thermal conductivity of Nb by considering the effect of phonon impurity scattering. This sample was heat treated at 800 °C for 2 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300 °C for 1 hour, at last it was heat treated again at 800 °C for 2 hours. The last heat treatment to degas the H impurity recovers the thermal conductivity at phonon peak.	 78
Figure 5.6:	Lattice thermal conductivity of Nb by considering the effect of phonon impurity scattering. This sample was heat treated at 140 $^{\circ}$ C for 48 hours and 1100 $^{\circ}$ C for 4 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300 $^{\circ}$ C for 1 hour, at last it was heat treated again at 800 $^{\circ}$ C for 2 hours. The last heat treatment has no effect to degas the H impurity and does not recover the thermal conductivity at phonon peak.	 79
Figure 6.1:	Phonon dispersion of Nb at [1 0 0] direction, replotted from the experimental data of [14]	 85
Figure 6.2:	Comparision between the calculation results and the data fitting results for an example sample of screw dislocations [4] for undeformed state and 8% tensile deformation. The group velocity is taken as $v_g = d\omega/dq$ , the dislocation density is calculated based on the fitting using Eq. 2.4.	 89

Figure 6.3:	Comparision between the calculation results and the data fitting results for another example sample of screw dislocations [4] for undeformed state and several levels of deformation. The group velocity is taken as $v_g = d\omega/dq$ , the dislocation density is calculated based on the fitting using Eq. 2.4	90
Figure 6.4:	Comparision between the calculation results and the data fitting results for an example sample of edge dislocations (sample H2) [4] for the undeformed state and 4% tensile deformation. The group velocity is taken as $v_g = d\omega/dq$ , the dislocation density was calculated based on the fitting using Eq. 2.4	91
Figure 6.5:	Comparision between the calculation results and the data fitting results for another example sample of screw dislocations (sample K10) [4]for the undeformed state and 1% and 2.4% tensile deformation. The group velocity is taken as $v_g = d\omega/dq$ , the dislocation density was calculated based on the fitting using Eq. 2.4.	92
Figure 7.1:	The thermal conductivity of Ta is the sum of the contributions of electrons and phonons [3]. Below 1 K, the electronic contribution is negligible. The contribution of phonons dominates in this temperature range because of the condensation of free electrons into Cooper pairs.	94
Figure 7.2:	Phonon dispersion relation of Ta in the [1 0 0] direction, replotted from [15], $\epsilon = aq/\pi$ , where q is the wave vector, $\epsilon$ is the reduced wave vector, a is the lattice parameter, which refers to the physical dimension of a unit cell in a crystal lattice. For Ta, $a \approx 3.3 \times 10^{-10}$ m, the same as Nb, LA refers to longitudinal acoustic phonons, and TA refers to transverse acoustic phonons	95
Figure 7.3:	Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 1 with the fitting results using Eq. 2.4 from [3], from undeformed to 2%, 3.1%, and 7.3% deformation	96
Figure 7.4:	Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 2 with the fitting results using Eq. 2.4 from [3], from undeformed to 0.5%, 1.2%, 2.4%, 3.9%, and 6.2% deformation	97
Figure 7.5:	Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 3 with the fitting results using Eq. 2.4 from [3], from undeformed to 1% deformation	98
Figure 7.6:	Comparison between fitting with the experimental data of the thermal con- ductivity of undeformed and deformed superconducting Nb [4] and Ta [16]. The dislocation density is calculated during the fitting processes.	99

Figure 7.7: Comparison between Monte Carlo simulation of the lattice thermal conductivity with the fitting results using Eq. 2.4 for undeformed Nb [4] and undeformed Ta [16]. The parameters, including size, RRR, deformation temperature, and the calculated dislocation density, of the Nb and Ta samples are listed in Table 4.1.100

Figure 7.8:	Comparison between Monte Carlo simulation of the lattice thermal conduc-
	tivity with the fitting results using Eq. 2.4 of undeformed and deformed Nb [4]
	and Ta [16]. The parameters, including size, RRR, deformation temperature,
	and the calculated dislocation density, of the Nb and Ta samples are listed in
	Table 4.1.

#### **CHAPTER 1**

### INTRODUCTION

Particle accelerators are increasingly used in a variety of applications for scientific research, medicine, and industrial processing. They accelerate beams of charged particles to high speeds (*e.g.*, high energy physics applications require particle beams to be accelerated to near the speed of light.) [17]. Radio frequency (RF) alternating current on the order of GHz provides energy to each charged particle for acceleration. Copper (Cu) was originally used for normally conducting cavities because of its easy formability and good electrical conductivity at room temperature. However, these cavities have high RF losses due to the finite electrical resistivity of normally conducting Cu, leading to loss of performance. Even with water cooling, Cu cavities cannot provide continuous operation because of the temperature excursions due to energy loss.

Alternatively, niobium (Nb) has been widely used to construct superconducting radio frequency (SRF) cavities in particle accelerators because it has the highest superconducting critical temperature ( $T_c = 9.25$  K) and the highest thermodynamic critical field ( $H_{th} = 1.6 \times 10^5$  A m<sup>-1</sup>) of any element [18]. As a superconductor, it has no electrical DC resistance and little RF resistance [5]. Manufacturing SRF cavities from large-grain Nb may reduce cost and improve the quality factor as compared with using polycrystalline Nb (*i.e.*, small-grain, typical grain size of about 50  $\mu$ m) [13], presumably because fewer grain boundaries exist in large-grain Nb. The resistance thus to decrease the performance of cavities. Even in the superconducting regime, residual resistance to RF energy and small imperfections at the RF surface dissipate energy and cause local heating that leads to cavity quench [19]. Large values of thermal conductivity can mitigate local temperature excursions by improving heat transfer to the liquid He bath, thus preventing cavity quench and improving cavity performance [20]. Processing Nb to obtain the largest thermal conductivity possible is an important strategy for improving performance.

Particle accelerators require the superconducting nature of Nb on the inner surface of a cavity



Figure 1.1: A typical example of the thermal conductivity of single crystal superconducting Nb sample from 0.3 K to 5 K with different levels of uniaxial strain, replotted from [4]. Undeformed state refers to Cauchy strain of 0%, and 1%, 2.4%, 4.7%, and 8.8% refer to the corresponding amounts of Cauchy strain. The thermal conductivity at temperature lower than 3 K decreases with the increase of strain. The phonon peak appears at 0%, 1%, and 2.4% deformation and disappears after 4.7% deformation. The thermal conductivity at the phonon peak decreases the most after deformation compared with the thermal conductivity at other temperatures.

to a depth of up to only the London penetration depth, which is about 30 nm for Nb [21]. This suggests that the superconducting properties of the cavity are only needed by a small fraction of the wall thickness (typically about 3 mm), which makes thin film superconducting composites an opportunity for the next generation of the cavities. The next generation of SRF accelerators may be developed using Nb clad Cu. This composite structure may take advantage of improved magnetic performance of thin film Nb along with the large values of thermal conductivity at low temperatures (*e.g.*, thermal conductivity of copper is on the order of magnitude of  $10^3$  Wm<sup>-1</sup>K<sup>-1</sup> at 2 K [22]).

The thermal conductivity of Nb at temperatures lower than  $T_c$  is significantly influenced by the processing history of Nb. A typical figure of thermal conductivity of Nb as a function of temperature is replotted from [4], shown in Fig. 1.1, at different levels of deformation. Purity and deformation have been demonstrated to change the thermal conductivity in both polycrystalline (small grain) and large grain Nb [7, 8]. Deformation is well known to reduce the local maximum thermal conductivity (also called the phonon peak  $k_{pp}$ ), but it is less known that it also increases the temperature at which the phonon peak occurs  $(T_{pp})$ . Several studies have shown that while plastic deformation may destroy the phonon peak, heat treatment may also restore it [8, 23]. The phonon peak of a single crystal Nb sample after 2.4% deformation has been measured to decrease by about 69% compared with its undeformed state [4]. The phonon peak temperature of the same specimen increased from 1.85 K at the undeformed state to 2.18 K after 2.4% deformation. Although the increase in temperature of the phonon peak is only 0.33 K, energy savings on this temperature difference could be significant for the working temperature of SRF cavity. This suggests that there is an optimization problem of balancing better performance of the cavities and more energy savings in the liquid He bath. Studying the mechanisms of both the thermal conductivity of bulk Nb and thin film Nb will provide guidance for the thermal design of current and next generation of SRF cavities.

This thesis examines the model of thermal conductivity of superconducting Nb buy including the effect of phonon-dislocation scattering. The relationship between  $k_{pp}$  and  $T_{pp}$  is found by examining different sets of measured data of the thermal conductivity of Nb. Fitting the data with the enhanced model leads to more accurate results than neglecting phonon dislocation scattering, especially at low temperatures(T < 3 K) and after deformation. The energy-based variance reduced Monte Carlo simulation technique is used to model and predict the lattice thermal conductivity of Nb before and after deformation using the relaxation time approximation. The simulation results match well with the fitting results using the enhanced model by considering the phonon-boundary scattering, phonon-electron scattering and phonon-dislocation scattering for both undeformed and deformed Nb samples.

### **CHAPTER 2**

### **REVIEW OF THERMAL CONDUCTIVITY OF NB**

Nb has great similarity to tantalum (Ta) in its physical and chemical properties, making them difficult to distinguish. As a refractory metal [24], Nb has a high melting point of 2477 °C which makes it resistant to wear, corrosion, and deformation. Nb has also been widely used as the material for superconducting cavities in particle accelerators because it has the highest superconducting critical temperature ( $T_c = 9.25$  K), the greatest thermodynamic critical field, and the greatest magnetic penetration depth [25] of any element. The superconductive properties, and especially the thermal conductivity are strongly dependent on the purity of Nb metal [26]. Very pure Nb may have high thermal conductivity at cryogenic temperatures, however, the high purity makes Nb metal comparatively soft and ductile [27], making it difficult to maintain dimensional stavility of SRF cavities. An example of a 9-cell SRF cavity developed at TESLA [5] is shown in Fig. 2.1.

The first measurements of the thermal conductivity of superconducting Nb focused on understanding the mechanisms of heat conduction in superconductors [28, 29]<sup>1</sup>. They compared the conductivity in the superconducting and normal conducting states at the same temperature by imposing a magnetic field to suppress the superconductivity of Nb below its critical temperature. Understanding the thermal conductivity of Nb took on a practical purpose as Nb became the material of choice for SRF particle accelerators starting in the 1970s [30]. SRF accelerators are typically comprised of thin-walled channels that are evacuated on the inside and submerged in liquid helium



Figure 2.1: An example of the sketch of a 9-cell TESLA accelerating structure [5]. The outside surface of the cavity is immersed into liquid He to keep Nb in superconducting temperature. Charged particles move inside the cavity to be accelerated by the magnetic filed.

<sup>&</sup>lt;sup>1</sup>Niobium was originally named columbium (Cb) and some earlier studies used that term.



Figure 2.2: Picture of the delamination crack. The crack is located approximately in the middle of the Nb sheet, spiting the sheet into two parts.



Figure 2.3: Microscopic image of the delamination crack. The crack propagates around 25 mm along the circumference, the thickness of the Nb sheet is around 2 mm, the crack splits the Nb sheet by half.

to operate in the temperature range of 1.8 K < T < 4.5 K. Oscillating electric fields accelerate charged particles along the axis of the evacuated channel to speeds that are a substantial fraction of the speed of light.

As a superconductor, Nb has no electrical resistance to direct current fields and small resistance to RF fields [5]. The resistance when subject to RF fields, as happens in a particle accelerator, leads to heat generation that must be dissipated to the helium bath surrounding the accelerator. This heat generation can lead to temperature excursions that may diminished accelerator performance to breakdown the magnetic field [20] and large values of thermal conductivity *k* at accelerator operating temperatures are essential for mitigating potential hot spots that would cause quenching of an accelerator cavity and degraded performance [5, 31, 32]. Thermal breakdown of an SRF cavity occurs when the RF surface temperature reaches its critical temperature [7]. Recent progresses in Nb cavities for particle accelerators has resulted in significant increase in thermal breakdown field up to  $180 \sim 200$  mT, which is close to the thermodynamic critical field (200 mT) [5, 32]. However, imperfections such as defects or dislocations can cause to a sharp decrease of thermal breakdown field [33]. Delamination cracks might also be a factor to decrease the field because of its resistance

to heat conduction between the RF surface and liquid He bath. Recent studies at FRIB of MSU [34] shows that after EB spot welding, a crack in the 2 mm Nb sheet of the outer conductor was discovered. The crack is approximately in the middle of the sheet, splitting the sheet in two parts and propagating around 25 mm along the circumference, the depth of crack cannot be determined. Fig.2.2 and Fig. 2.3 show macro the picture and microscopic images of the delamination crack, respectively.

Padamsee correlated the peak magnetic field with the purity of Nb as characterized by its electrical resistivity ratio (RRR) [35].

$$RRR = \frac{\rho_{295K}}{\rho_{4.2K}} \tag{2.1}$$

where  $\rho_{295K}$  and  $\rho_{4.2K}$  are the electrical resistivity at 295 K and 4.2 K, respectively.

The thermal conductivity at 4.2 K of superconducting Nb can also be correlated to RRR [36] as in Eq. 2.2. This illustrates the importance of having a large value of thermal conductivity.

$$RRR = 4k_{4.2K} \tag{2.2}$$

where  $k_{4.2K}$  is the thermal conductivity at the temperature of 4.2 K.

Heat conduction in metals occurs by conduction of normal conducting electrons and lattice vibrations (*i.e.*, phonons) [37]. These two components are additive [38] such that

$$k = k_e + k_g \tag{2.3}$$

where  $k_e$  represents heat conduction by electrons and  $k_g$  represents the heat conduction by phonons. In normal conducting metals, the electron component dominates and the phonon part is usually negligible. In the superconducting regime, however, the condensation of normal conducting electrons into Cooper pairs leads to a reduction in the electron contribution to energy transport, as well as an increase in the phonon contribution due to reduced scattering of phonons by electrons [39]. Therefore, the phonon component of heat conduction increases in significance in superconducting materials.



Figure 2.4: The contributions of the normal electron thermal conductivity  $k_e$  and the lattice thermal conductivity  $k_g$  to the overall thermal conductivity  $k = k_e + k_g$ . At temperature lower than 2 K,  $k_g$  dominates the total thermal conductivity and  $k_e$  dominates the thermal conductivity for temperature higher than 3 K. For this Nb with relatively few dislocations, there is a significant phonon peak.

Representative contributions of  $k_e$  and  $k_g$  for superconducting Nb are shown in Figure 2.4. The Nb used for illustration has relatively few dislocations, such as a material that has been relatively undeformed after a proper heat treatment. For  $T \ge 3$  K,  $k_e > k_g$  and the contribution of  $k_g$  decreases with increasing T as phonons are increasingly scattered by normal conducting electrons. For  $T \le 2$  K,  $k_e$  is negligible as essentially all of the electrons have formed Cooper pairs [39]. As a result of the reduced scattering of phonons, a phonon peak in the thermal conductivity  $k_{pp}$  is evident at  $T \approx 2$  K. As the Nb cools below 2 K,  $k_g$  decreases due to boundary scattering until vanishing at 0 K.

Experimental illustration of the electrical and lattice contributions to k is given in Fig. 2.5,

which compares six tests on three specimens from three studies [4, 6, 7]. Others have shown similar responses using other measurements (e.g., [40]). The red circles are for a Nb specimen with RRR = 1200 [4]. The open circles are for Nb that is undeformed following annealing and the closed symbols are for the same sample having undergone a uniaxial Cauchy strain of  $\epsilon = 8\%$ . While the conductivity for T > 3 K is the same before and after deformation of  $\epsilon = 8.5\%$ , the  $k_{pp}$ at  $T \approx 2$  K disappears after deformation, likely due to the increase in dislocation density. It is worth noting that  $k \approx 175$  W m<sup>-1</sup> K<sup>-1</sup> at T = 4.2 K in this specimen, instead of the expected k = 300 W  $m^{-1}$  K<sup>-1</sup>, which does not agree with Eq. 2.2. The solid black squares are for Nb with RRR = 390 in its "as-received" condition (i.e., with unknown deformation) [7]. The thermal conductivity for T > 3 K behaves as expected, and there is no  $k_{pp}$ . The open squares are for the same specimen after it had been annealed, although inadvertently without a titanium getter. Without a getter, the impurity concentration increased, as exemplified by the RRR decreasing to 68. This suggests that although the RRR decreased, the dislocation density had also decreased with the result of increasing k for  $T \le 3$  K. The triangles in Fig. 2.5 are for a Nb specimen with RRR = 255 before deformation and RRR = 237 after deformation, a small change as exemplified by k for T > 3 K [6]. The open triangles are for the undeformed specimen and filled symbols are for the Nb after deformation to  $\epsilon = 8.5\%$ . The undeformed specimen has a phonon peak at about 2 K. After the specimen was strained to a Cauchy strain of  $\epsilon = 8\%$ , there is no  $k_{pp}$ .

Given the practical and fundamental importance of k in superconducting Nb, it has been subject to numerous studies. Normal conducting electrons largely determine k for  $T_c/3 \leq T \leq T_c$ , even as they condense into Cooper pairs. At these temperatures, the impurity concentration largely determines k. As the temperature decreases further, the paired electrons no longer conduct thermal energy and no longer scatter phonons. The lack of electron scattering, can lead to a local maximum in k, that is  $k_{pp}$ . But other mechanisms scatter the phonons, including material boundaries, dislocations, and grain boundaries. Other effects can greatly alter the thermal response of Nb. Magnetic fields, for example, can suppress the superconductivity in type II superconductors, and leave the normal conducting electrons as the dominant mode of energy transport. The following



Figure 2.5: Illustration of the roles of dislocation density and RRR on polycrystalline [6, 7] and single crystal Nb [4]. Thermal conductivity at temperature T=4.2 K is proportional to the RRR. However, at the temperatures close to 2 K, the strain has more effect on the thermal conductivity than the RRR. Of particular note is that a single sample is measured in its as-received state at RRR=390, after annealing at 1200 °C for two hours, but with a Ti getter, the purity is reduced to RRR=68, but a phonon peak develops.

sections review the studies that have measured thermal conductivity in superconducting Nb subject to many external effects.

## **2.1** Experiments on thermal conductivity of Nb

To put the measurements into context, it is instructive to examine a model for k that is based on Bardeen-Cooper-Schrieffer (BCS) theory [39]. Koechlin and Bonin [41] developed an expression for k of superconducting Nb by parameterizing the Bardeen-Rickayzen-Tewordt (BRT) [42] model for k based on BCS theory [39]. Xu *et al.* [43] added a term to the Koechlin and Bonin model to account explicitly for phonons scattered by dislocations in the lattice thermal conductivity. This augmented model can be written as

$$k = R(y) \left[\frac{\rho}{LT} + AT^2\right]^{-1} + \left[\frac{1}{D\exp(y)T^2} + \frac{1}{B\Lambda T^3} + \frac{KN_d}{T^2}\right]^{-1}$$
(2.4)

where the two terms in the first brackets account for electrons scattered by impurities and electrons scattered by phonons, respectively, and the three terms in the second brackets account for phonons scattered by electrons, phonons scattered by boundaries, and phonons scattered by dislocations, respectively. The term R(y) quantifies the condensation of normal conducting electrons into Cooper pairs,  $\rho$  is the residual electrical resistivity ( $\rho = \rho_{295\text{K}}/\text{RRR}$ ), RRR is the residual resistivity ratio,  $\rho_{295\text{K}}$  is the electrical resistivity at 295 K,  $L \approx 2.45 \times 10^{-8}$  W K<sup>-2</sup> is the Lorentz number, *A* is the coefficient of momentum exchange of electrons with the lattice, *D* scales phonon-electron scattering, *B* scales phonon-boundary scattering,  $\Lambda$  is the phonon mean free path,  $N_d$  is the dislocation density and *K* is a parameter describing phonon-dislocation scattering. The term R(y) may be calculated as

$$R(y) = \frac{k_{es}}{k_{en}} = f(0)^{-1} \left[ f(-y) + y \ln(1 + \exp(-y)) + \frac{y^2}{(2(1 + \exp(y)))} \right]$$
(2.5)

where  $k_{es}$  and  $k_{en}$  are the electronic thermal conductivity of superconducting state and normal conducting state, respectively. The ratio of the superconducting energy gap ( $\Delta(T)$ ) to thermal fluctuations of  $k_BT$  may be written as

$$y = \frac{\Delta(T)}{k_B T} = \frac{\Delta(T)}{k_B T_c} \frac{T_c}{T}$$
(2.6)

and f(-y) as

$$f(-y) = \int_0^\infty \frac{z dz}{1 + \exp(z + y)}$$
(2.7)

For  $T/T_c < 0.6$ , y can be approximated as  $y = \alpha T_c/T$  with  $\alpha \approx 1.76$  from BCS theory. The parameters  $\rho$ , *a*, *B*, *D*,  $N_d$ , and  $\alpha$  are typically estimated from measurements of conductivity. The parameter *K* is expressed following Klemens [44] for randomly distributed dislocations as

$$K = \frac{0.038(\bar{v}h^2)b^2\gamma^2}{k_B{}^3}$$
(2.8)

where  $\gamma$  is the Grüneisen constant ( $\gamma$ =1.4 for Nb), *b* is the Burgers vector,  $\bar{\nu}$  is the average group velocity of Nb, and *h* is the Planck constant. Using values for Nb, an expression for  $K = 3.05 \times 10^{-15}$  m<sup>3</sup>K<sup>3</sup> W<sup>-1</sup>.

The expression by Koechlin and Bonin [41] has been used successfully to model the thermal conductivity of superconducting Nb in many circumstances. The extended expression of Eq. 2.4 shows promise for improving modeling of *k* for superconducting Nb and Ta with substantial dislocations, especially at  $T/T_c \approx 0.22$  [43].

To our best understanding, the first measurement on the thermal conductivity of Nb is conducted by Mendelssohn and Olsen at 1950 [28]. They measured the thermal conductivity of Nb rod from superconducting temperature to normal conducting temperature and found that the thermal conductivity curve behaved differently than other superconductors (Ta and Pb-Sn alloys) as *k* at superconducting state exceeds the values at normal conducting state for temperatures below 4 K. They explained this feature to be the reason of appearance of circulation heat flow at low temperatures because of the applied magnetic flux. Two years later, Mendelssohn and Rosenberg [29] measured the thermal conductivity of 99.99% pure Nb at temperatures from 2 K to 40 K. They showed similar behavior with reference [28] as the thermal conductivity curve at superconducting and normal conducting state (because of the applied magnetic flux) crossed over with each other for the magnetic field of 2300 & 3300 Gauss field at T < 6 K and T < 3 K, respectively. The reason is that the decrease of lattice *k* because of the scattering of phonons by extra electrons produced by the magnetic field was not fully compensated by the increase of electronic *k*. Mendelssohn [45] investigated the thermal conductivity of several superconductors experimentally below 1 K. He obtained the thermal conductivity of poly-crystal Nb to be proportional to  $T^3$  between 0.4 K and 0.75 K, which agrees with Casimir model [46] for boundary scattering of dielectric materials. He thus concluded that the thermal conductivity of superconductive metal near absolute zero behaves like that of a dielectric crystal. The Casimir model [46] for the lattice thermal conductivity due to crystal boundary scattering was expressed as

$$k_b = 2.31 \times 10^3 RPA^2 / 3T^3 \tag{2.9}$$

where *R* is the radius for a circular cylindrical specimen,  $A = C_v/T^3$  with  $C_v$  being the specific heat per unit volume, *P* is expressed as

$$P = \frac{2(v_l/v_t)^2 + 1}{[2(v_l/v_t)^3 + 1]^2/3}$$
(2.10)

where  $v_l$  and  $v_t$  are the longitudinal and transverse velocities of sound in the material, respectively.

In 1958, Mendelssohn [47] was the first to quantify the phonon conduction in superconductors to the number of dislocations by measuring the thermal conductivity of superconducting lead and niobium. Van Bueren's formula [48] was used to calculate the dislocation density for face centered cubic (FCC) lattices, (*i.e.*, lead). For body centered cubic (BCC) materials, (*i.e.*, Nb), Klemens model [44] was used to estimate the dislocation density. Mendelssohn [47] also mentioned that the superconductive metal can be rendered normal by applying a low magnetic field, and the strain only affects the thermal conductivity of the superconducting state, not the normal state. Another observation in this paper is that the zone refined Nb sample showed a maximum in thermal conductivity in the superconducting state that exceeded that of the normal state at 2 K. After zone refining process, the thermal conductivity was measured to be  $180 \text{ Wm}^{-1}\text{K}^{-1}$  at 2 K, whereas before zone refined process  $k_{pp} \approx 75 \text{ Wm}^{-1}\text{K}^{-1}$  at 2.4 K, which was less than the thermal conductivity in the normal state.

Montgomery [49] measured the thermal conductivity of lead between 1 K and 4 K and noted that below about 1.4 K, the thermal current is carried entirely by lattice waves, but not conduction

electrons, which is consistent with [45]. He also found that grain boundaries are responsible only for a part of the phonon scattering, with sub-grain structure and dislocation networks being of comparable importance. He suggested that thermal conductivity of superconductors at superconducting state becomes size dependent at low temperatures. Chaudhuri et al. [50] measured the thermal conductivity of Nb and V at superconducting temperatures and investigated the effect of radiation on the thermal conductivity. The effect of neutron irradiation on the thermal conductivity of superconducting Nb behaves similarly as dislocation scattering to have the thermal resistance W being proportional to  $T^{-2}$ . The explanation is that neutron irradiation introduces interstitials in the sample, where the condensation of interstitials increases the dislocations. In 1961, Mendelssohn [51] examined different aspects of the thermal conductivity of metals at low temperatures, *i.e.*, impurity, strain, and irradiation. Electrons are mainly scattered by impurities [44], therefore they affect the thermal conductivity in both normal and superconducting state. Phonons are scattered by dislocations, often formed by plastic deformation, and contribute to the lattice thermal conductivity. Therefore, strain only affects the thermal conductivity in the superconducting state. The effect of neutron irradiation on the thermal conductivity of superconducting Nb behaves like phonondislocation scattering. However, in the normal state thermal conductivity can also be reduced by neutron irradiation, because vacancies were produced by irradiation [50].

Calverley *et al.* [52] systematically investigated the thermal and magnetic properties of Ta, Nb, and V at liquid He temperatures. For the effect of strain on thermal conductivity of Nb, they observed reduced thermal conductivity following deformation. Their figures of thermal conductivity show that the phonon peak temperature gradually increased with deformation until the peak disappeared because of increased dislocation density. Connolly and Mendelssohn [53] measured the thermal conductivity of Nb between 0.2 K and 4.2 K. They found a local maximum at about 0.2  $T_c$  (T=1.85 K) and analyzed the electron and lattice components of k. They concluded that contribution of phonon conduction needs to be considered for  $T < 0.5T_c$ . Kuhn [54] measured the thermal conductivity of Nb and investigated the effect of irradiation, phonons scattered by dislocations, and phonons scattered by electrons using the BRT [42] model. Thermal conductivity in the superconducting state can exceed that of the normal conducting state for Nb after recrystalization. Wasim and Zebouni [55] measured the thermal conductivity of Nb of intermediate purity  $(l/\xi_0 \approx 1)$ , where *l* is the electronic mean free path, and  $\xi_0$  is the coherence length) in the mixed states. The clean and dirty limits of superconductors are defined as the comparison between *l* and  $\xi_0$ . If  $l \ll \xi_0$ , it is referred as dirty superconductors [56], otherwise, it is referred as clean superconductors. For dirty superconductors, the impurities are usually high that can scatter with electrons to reduce the thermal conductivity, both phonon and electron thermal conductivities decrease just above the lower critical field  $(H_{c1})$ .

As a type-II superconductor, Nb has two critical magnetic fields, namely  $H_{c1}$  and  $H_{c2}$  [5]. When the applied magnetic DC magnetic field exceeds  $H_{c1}$ , the normal conducting magnetic flux vortices penetrate the Nb surface. However, the Nb material around these vortices is still in superconducting state. When the applied magnetic DC magnetic field is greater than  $H_{c2}$ , the magnetic flux vortices penetrate the Nb surface completely and transform the superconductor into normal conductor. When the applied magnetic field lies between  $H_{c1}$  and  $H_{c2}$ , this is the mixed state. For Nb at 0 K,  $H_{c1} = 1.08 \times 10^4$  A/m,  $H_{c2} = 1.91 \times 10^4$  A/m [5].

Mittag [57] measured the Kapitza resistance and thermal conductivity of Nb from 1.3 K to 2.1 K, the Kapitza resistance is higher for a sample after annealing and chemical polishing than for a sample that was only machined. Carlson and Satterthwaite [58] measured the thermal conductivity of Nb between 0.3 K and 5 K. They found an anomalously high thermal conductivity (a factor of 5 higher than BCS) between 0.3 K and 0.6 K and proposed it was due to a small second energy gap. However, Anderson *et al.*, [59] measured the thermal conductivity of single-crystal Nb from 0.04 K to 4 K and found no evidence of a second energy gap. From another measurement of four single-crystal Nb samples, Anderson and Smith [23] proposed a resonant like scattering of thermal phonons lying near  $5 \times 10^{10}$  Hz, which is independent of stain and impurity. They explained this feature to be a dynamic phonon-dislocation interaction. In 1974, Anderson and O'Hara [60] studied the lattice thermal conductivity of Nb as limited by the interaction of phonons with electrons. They indicated that the relationship between the mean free path of transverse and

longitudinal acoustic phonons and temperature are similar, both are about  $4 \times 10^{-5}T^{-1}$ . Kes *et al.* [61] measured the thermal conductivity of Nb considering crystal defect structures like dislocations, grain boundaries, and dislocation loops due to neutron irradiation. They studied different scattering mechanisms with relaxation time approximation, including grain boundary scattering, point defect scattering, dislocation scattering, and phonon electron scattering, based on BRT theory [42] and Klemens model [44]. They found that dislocation loops introduced by neutron irradiation behave like clusters of point defects, and irradiation can enhance the original dislocation term. The relaxation times used in this paper are expressed as the following.

Grain boundary scattering:

$$\tau_{GB}^{-1} = 0.03\gamma^2 \alpha^2 v_s N_g \tag{2.11}$$

where the Grüneisen constant has been defined in Eq. 2.8,  $\alpha$  is the angle of tilt of the grain boundary ( $\alpha < \pi/4$ ),  $v_s$  is the sound velocity, and  $N_g$  is the number of grain boundaries crossing a line of unit length.

Strain field scattering:

$$\tau_D^{-1} = 0.033 \gamma^2 b^2 N_d \omega \tag{2.12}$$

where  $\omega$  is the phonon frequency.

Point defect scattering:

$$\tau_p^{-1} = \frac{3a^3 S^2 \omega^4}{G\pi v_s^3} \tag{2.13}$$

where  $a^3$  is the volume per atom, G is the number of items per crystal, and S is the scattering amplitude of the point defects. For the difference in mass of the impurities  $S_1^2 = (\Delta M/M)^2/12$ , for the lattice distortion by the impurities,  $S_2^2 = 3\gamma^2(\Delta R/R)^2$ , where *R* is the lattice spacing.

Phonons scattered by electrons:

$$\tau_{pe}^{-1} = g(x, T)ExT$$
 (2.14)

where  $x = \frac{\hbar\omega}{k_BT}$ , with  $\hbar$  being the reduced Planck constant,  $k_B$  being the Boltzmann constant. The term g(x, T) is a monotonically decreasing function of x, and E is a fitting parameter.

Moore and Satterthwaite [62] measured the thermal conductivity of pure Nb in magnetic field near upper critical field  $H_{c2}$ . Measurement results confirmed the theory that the mixed state thermal conductivity varied with field at the magnetic field near  $Hc_2$ , which was also verified by Kes et al. [63] by measuring the thermal conductivity of Nb in the mixed state at different field orientations. Mamyia et al. [64] measured the thermal conductivity of Nb with different RRR of 6300, 2500, and 195. Niobium, as an intermediate coupling superconductor, behaves differently to the electronic thermal conductivity due to electrons scattered by phonons from strong coupling superconductors (e.g. Lead) and weak coupling superconductors (e.g. Tin). They characterized the slope of the phonon scattering term of reduced thermal conductivity against reduced temperature for intermediate coupling superconductor Nb to be 2.8, as compared with 1.5 for weak coupling superconductors. Here the reduced thermal conductivity is defined as  $k_S/k_N$  where  $k_S$  is the thermal conductivity at superconducting state and  $k_N$  is the thermal conductivity at normal conducting state. And the reduced temperature is defined as  $T/T_c$ . Gladun *et al.* [65] measured the thermal conductivity of single-crystal Nb of high purity (RRR=33000<sup>2</sup>) at the temperature range from 0.05 K to 23 K. They found the thermal conductivity values in the range of six orders of magnitude  $(k = 2 \times 10^{-3} \text{W m}^{-1} \text{ K}^{-1} \text{at } 0.05 \text{ K to } k = 2.2 \times 10^{3} \text{W m}^{-1} \text{ K}^{-1} \text{at } 9 \text{ K})$ . The surface roughness of the sandblasted sample was characterized and they concluded that the difference in thermal conductivity of the sandblasted sample below 2 K may be due to the annealing of dislocations in the surface layer or within the crystal due to sandblasting. Reiche and Pompe [66] investigated the thermal conductivity and electrical conductivity of bulk niobium and niobium films in the normal state from 9 to 50 K, based on the deviations from the Matthiessen rule. They showed a slight dependence of the electron-phonon scattering coefficient on the impurity content of the sample. An additional resistance term proportional to  $T^{5.5}$  was proposed in the temperature range from 20 to 50 K.

 $<sup>^{2}</sup>$ RRR=33000 is the theoretical limit for Nb



Figure 2.6: A typical example of the thermal conductivity of single crystal superconducting Nb sample with screw dislocations from 0.3 K to 5 K with different levels of uniaxial strain, replotted from [4]. Undeformed state refers to 0 Cauchy strain, and 3%, 7.3%, and 14.7% deformation correspond to the same amount of Cauchy strain. The thermal conductivity at temperature lower than 3 K decreases with the increase of strain. The phonon peak appears at 0% deformation and 3% at about 1.8 K and 2 K separately. There is nearly no phonon peak after 7.3% deformation and 14.7% deformation. The thermal conductivity data at temperatures higher than 3.5 K are the same for different amount of deformations.

Wasserbäch systematically measured the thermal conductivity of single-crystal Nb and Ta in the temperature range  $0.3 \le T \le 20$  K. He considered the effects of interstitial and substitutional impurities [67], plastic deformation [4], phonons scattered by electrons [68], deformation at intermediate temperatures [3], and heterogeneity of the dislocation distribution [69] on the lattice thermal conductivity of Nb. The thermal conductivity of Nb below 1 K was found proportion to  $T^3$ , but its magnitude decreased with increasing impurity content [67]. In 1978, Wasserbäch measured the effect of the temperature at which deformation occurs on the thermal conductivity of Nb specimens with different RRR values. He characterized the deformation as weak ( $\epsilon \leq 2\%$ ), medium  $(2\% \le \epsilon \le 6\%)$  and strong  $(\epsilon \ge 7\%)$ , although these varied somewhat. Figure 2.6, similar to Fig. 1.1, shows the typical response of thermal conductivity of Nb to temperature and deformation. For weak and medium deformation at 77 K and 195 K, the thermal resistance of phonons scattered by dislocations was proportional to  $T^{-3}$ . These dislocations were measured by transmission electron micrographs (TEM) to be predominantly screw dislocations. Strong deformation at 195 K and medium to strong deformation at 295 K and 470 K gave rise to thermal resistance proportional to  $T^{-2}$ . Deformation at 295 K and 470 K led to predominantly edge dislocations. Experimental investigation of phonons scattered by electrons [68] indicated that both the longitudinal and transverse acoustic phonons were scattered strongly by electrons for Nb, while only longitudinal acoustic phonons are scattered by electrons in Pippard's theory [70]. Wasserbäch's results agree with the phonon mean free path investigation by Anderson et al. [60]. Single crystal Nb or Ta plastically deformed at T = 355, 370, or 420 K demonstrated thermal resistance proportional to  $T^{-2}$ [69], which is consistent with [4] and with Klemens's model [44] of phonon scattering by a static distortion field of dislocations.

Williams *et al.* [71] measured the thermal conductivity and electrical conductivity of Nb and Nb-based alloys in the temperature range 80-400 K. They analyzed the data by considering phonon-phonon and phonon-electron scattering in limiting the phonon thermal conductivity and compared the phonon-electron thermal resistance with first-principles calculation with good agreement. Krafft [72] measured the surface resistance, thermal conductivity and Kapitza resistance between Nb and

liquid He and showed that the above three parameters must be established to make predictions about the role of thermal transport in determining the performance of SRF cavity. He also mentioned that high thermal conductivity did stablize the cavity surface against local heating due to defects. Padamsee [35] studied the influence of thermal conductivity on the breakdown field of Nb cavities by varying the RRR values in 40 different tests. He found that the breakdown field of Nb cavity is proportional to the square root of the RRR. A new purification technique [73] was used to improve the thermal conductivity of Nb by removing the dominant interstitial impurity (oxygen) in Nb. Hörmann [74] described the manufacturing process for superconducting niobium. He mentioned that electron beam melting and hot and cold working may increase the commercial Nb RRR from 20-40 to 160-300, and the thermal conductivity from 8-10 W m<sup>-1</sup> K<sup>-1</sup> to around 70 W m<sup>-1</sup> K<sup>-1</sup>. Kneisel et al. [75, 76] investigated the SRF performance of high field cavities by comparing material properties such as thermal conductivity, residual resistivity, and tensile behavior. Koechlin et al. [77] measured the thermal conductivity of Nb from 1.5 to 20 K. They concluded that high purity contributes to good thermal conductivity by electrons when T > 2.5 - 3K, however, low thermal conductivity in the temperature range of 1.8-2.0 K is due to small grain dimensions. Moderate temperature annealing (800 °C < T < 1200 °C) can increase the grain size from 50-70  $\mu m$  to 400-500 µm and the thermal conductivity at around 2 K. Boucheffa et al. [78] measured the Kapitza resistance and thermal conductivity of Nb. They showed that thermal conductivity at T > 3 K increases with RRR, however for T < 3 K, the thermal conductivity behavior becomes dependent on other factors, particularly the grain size. Also, the Kapitza resistance of RRR $\approx 200$  is strongly dependent on its surface treatment. Ikebe [79] measured the thermal conductivity of Nb based alloys and fitted the data with the Tewordt and Wölkhausen (TW model) [80] with good agreement.

Amrit [81] measured the thermal conductivity of poly-crystal Nb and investigated the effect of phonon-grain boundary scattering on the thermal conductivity. He showed that the thermal conductivity decreases strongly as the number of grains increase up to a limit of 10. When the number of grains exceeds 10, the anisotropy of thermal conductivity disappears. Singer *et al.* [13] investigated the SRF performance of large grain cavities using both the basic material properties and material availability, production, and preparation aspects. They obtained a gradient up to 41 MV/m at the quality factor  $Q_0 = 1.4 \times 10^{10}$ .

Gurevich [32] studied the effect of nonlinear surface resistance on the SRF thermal breakdown and proposed a model of non-uniform thermal breakdown by macroscopic hotspots on the cavity surface. Aizaz et al. [7] measured the thermal conductivity and Kapitaza conductance of Nb and studied the effect of plastic deformation on phonon transmission and Kapitza resistance. They found the absence of phonon peak and the reduced Kapitza conductance are due to the applied strain. Low temperature annealing (around 750 °C) did not restore the phonon peak. Kneisel et al. [82] tested six single cell Nb cavities at TESLA. After appropriate surface treatments by buffered chemical polishing and eletropolishing, they are able to reach the accelerating gradients between 38 MV/m and 45 MV/m. Singer *et al.* [31] measured the RRR of Nb using DC and AC methods and analyzed the advantages and disadvantages of these two methods. Basically, the DC method has higher accuracy but is a destructive method only good for quality control of Nb before start of cavity fabrication. The AC method preformed directly on the cavity surface and makes it possible to estimate the RRR value, however, the accuracy is less than that of DC method. Aizaz et al. [83] studied the thermal designs in SRF Nb cavities by considering the phonon peak and Kapitza conductance. Strain can reduce the thermal conductivity at its phonon transmission regime by 80%. Although low temperature annealing did not restore the phonon peak [7], moderate temperature (around 1300 °C) annealing during titanification process can restore the phonon peak in the thermal conductivity curve.

Chandrasekaran *et al.* [8, 84, 85] measured the thermal conductivity of bi-crystal Nb with different RRR values and considered the influence of grain size and grain orientation, impurity, deformation, and heat treatment on the thermal conductivity of Nb. They proposed a novel parameter estimation method [86] based on temperature and heat flux measurements. The phonon peak was found at close to 1.8 K [8] for nearly of all the undeformed samples. While plastic deformation has the ability to destroy the phonon peak [8], heat treatment with appropriate temperature and duration may restore it. Dhakal *et al.* [12] measured the thermal conductivity of single crystal

Nb and analyzed the parameters by fitting with the model proposed by Koechlin and Bonin [41]. Chandra *et al.* [87] investigated the magnetic field dependence of the thermal conductivity of Nb. They found that the quasi-particle scattering from vortices dominates in the low field, however, the intervortex quasi-particle tunneling dominated in high fields. Mondal *et al.* [88] measured the thermal conductivity of large grain niobium and investigated its dependence on trapped vortices in a parallel magnetic field. They found that the phonon peak at around 2 K disappeared due to the presence of trapped vortices because of strong scattering of phonons by vortex cores.

To the best of our knowledge, the details of researchers' contributions to experiments on thermal conductivity of Nb are listed in Table 2.1.

Table 2.1: Chronology of measurements of the thermal conductivity of superconducting Nb. RRR. values marked with asterisks (\*) are estimated using measured  $k(4.2 \text{ K}) \times 4$ 

Year	RRR	<i>T</i> (K)	Contributions	Ref.	
1950	99.99%	2-22	w/ & w/o magnetic field	[28]	
1952	99.99%	2-22	w/ & w/o magnetic field	[29]	
1955	99.99%	0.4-0.75	$k \propto T^3$ with magnetic shielding	[45]	
1958	36*	1-4.2	Dislocation, $\epsilon = 0, 5\%, 13.4\%, 19.5\%$	[47]	
1960	48-54*	1-4.2	Fast neutron irradiation	[50]	
1961	52*	0.4-9	Dislocation, irradiation, RRR, and boundary	[51]	
1960	400*	1-4.2	Plastic deformation & dislocation density	[89]	
1961	56-40*	1-4.2	Measurements in single- and poly-crystal sample	[52]	
1962	60.5-120	0.2-4.2	Electronic and lattice contribution	[53]	
1966	NA	0-9.25	Measurements in normal and superconducting metals	[54]	
1969	29	1.95-9.0	k at mixed state of intermediate purity	[55]	
1970	196	0.3-6	Anomalous k for $T \le 0.6$ , possible 2 <sup>nd</sup> energy gap	[58]	
1971	99.9%	1.3-2.1	$h_{\text{Kapitza}}$ , annealed and chemically polished	[57]	
	Table 2.1 (cont'd)				

				,
Year	RRR	<i>T</i> (K)	Contributions	Ref.
1971	26 & 2000	0.04-4.2	No evidence of 2 <sup>nd</sup> energy gap	[59]
1972	NA	1.79-2.0	Magnetic fields near $H_{C2}$ , electron mean free path	[62]
1973	261-961	0.03-4	Dislocation, resonant phonon scattering	[90]
1973	NA	0-14	Neutron scattering measurement of phonon line width	[91]
1974	26-1800	0.5-4	$k_g$ , longitudinal and transverse phonons	[60]
1974	100	0.1-4.2	Hydrogen impurity	[92]
1974	15.6-24.3	1-10	Dislocation, irradiation, and grain boundary	[61]
1974	195	1-15	Phonon scattering of inter. coupling superconductors	[64]
1975	15.6-24.3	2.13-6.04	Magnetic field orientation, mixed state	[63]
1977	8000*	0.05-23	Surface roughness, phonon scattering	[65]
1977	300	0.3-20	Interstitial and substitutional impurity	[67]
1978	185-1200	0.3-20	Phonon-dislocation scattering	[4]
1979	11.4-4910	9-50	Bulk Nb and Nb thin films	[66]
1983	NA	80-400	$k_g$ at intermediate T	[71]
1983	500	1.5-4	Thermal transport of metal Kapitza resistance	[72]
1985	25-2000	1-9	k on thermal breakdown field	[35]
1985	65-300	4-9	New purification technique to improve $k$	[73]
1985	185-1200	0.3-20	Electron-phonon scattering	[68]
1985	185	0.3-20	Deformation at intermediate temperatures	[3]
1987	185-1200	0.3-20	Heterogeneity of dislocation distribution	[69]
1988	25-350	2-10	Electron beam melting, hot and cold working	[74]
1991	40-250	1.5-20	Anneal, grain size, RRR	[77]
1994	38-270	1.4-2.15	$h_{\text{Kapitiza}}$ and $k$	[78]
1996	N.A.	1.2-4.2	Lattice k of Nb-Ta alloys	[79]
Table 2.1 (cont'd)				

Table 2.1(cont'd)
Year	RRR	<i>T</i> (K)	Contributions	Ref.
2002	2840-10500	0.03-3	Internal friction	[93]
2006	673	1.6-2.1	Number of grains, grain boundary $h_{\text{Kapitiza}}$	[81]
2007	200-500	$0-T_c$	Large grain Nb performance	[6]
2007	230-300	1.6-4.2	Plastic deform., low T heat treatment, $h_{\text{Kapitiza}}$	[7]
2008	280,470	$0-T_c$	Single crystal Nb, SRF performance	[82]
2010	230-300	1.6-4.2	Phonon peak, $h_{\text{Kapitiza}}$	[83]
2011	104-196	1.5-4.5	Heat treatment, phonon scattering	[85]
2012	224±8	$1.5 - T_c$	BCP, heat treatment, surface resistance	[12]
2012	280	2-12	Magnetic field, impurity	[87]
2012	61-158	1.8-5	Trapped vortice, large grain Nb	[88]
2013	104-196	1.5-4.5	Grain size, orientation, impurity, $\epsilon$ , H.T., $k_{pp}$	[8]
2017	N.A.	0.1-0.6	Thin wires, $k$ at low $T$ deviates from BCS	[94]

Table 2.1(cont'd)

# 2.2 Models on thermal conductivity of Nb

Heat is carried by both the conduction electrons and the lattice vibrations in metals [37]. The thermal conductivity of Nb can be modeled as consisting of electron transport of energy and phonon transport of energy. These two components are additive and can be expressed as Eq. 2.3. In normally conducting metals, the phonon part is usually negligible due to scattering of normal electrons. However, in the superconducting regime, the formation of free electrons into Cooper pairs leads to a reduction in the electron contribution to energy transport as well as a reduction in scattering of phonons by electrons [39]. Therefore, the phonon contribution to thermal conduction increases in significance in superconducting materials.

Callaway [95] developed a phenomenological model for the lattice thermal conductivity at low temperature by considering the different scattering mechanisms of phonons, such as boundary

scattering, the normal three phonon process, impurity scattering, and Umklapp process, based on the Boltzmann transport equation (BTE). At very low temperatures, the boundary scattering is dominant, the normal process is small and can be neglected. The total lattice thermal conductivity can be expressed as

$$k = \frac{k_B^3}{\hbar^3} \frac{k_B}{2\pi^2 v_g} T^3 \int_0^{\Theta_D/T} \frac{\tau \exp(x) x^4}{(\exp(x) - 1)^2} dx$$
(2.15)

where  $x = \frac{\hbar\omega}{k_BT}$  is the non-dimensional frequency,  $v_g$  is the group velocity, here taken as a constant with the value of sound velocity (no dispersion considered),  $\tau$  is the overall relaxation time of different scatterings, including phonon-phonon scattering, phonon-electron scattering, phonon-impurity scattering, phonon-boundary scattering (phonon-grain boundary scattering), and phonon-dislocation scattering, and  $\Theta_D$  is the Debye, here taken as  $\Theta_D=275$  K for Nb.

The Callaway model assumes that all phonon modes are scattered independently of one another and uses the Debye approximation. The Debye approximation assumes that there is no phonon dispersion, in other words, the longitudinal and transverse acoustic polarizations behave identically. Holland [96] extended Callaway's model by treating transverse and longitudinal acoustic phonons differently to estimate the lattice thermal conductivity. This model agrees well with the Callaway model in the impurity scattering and boundary scattering regions. The Holland model improves the accuracy of estimating the lattice thermal conductivity by considering the linear dispersion. However, this model still considers the overall relaxation time. Moreover, as the model is under the assumption of linear dispersion, it would be difficult to estimate the lattice thermal conductivity of materials with non-linear dispersion.

Klemens [44] did systematic research on the expressions of scattering relaxation times which contribute to the lattice thermal conductivity, including phonon-phonon scattering, phonon-impurity scattering [44], phonon-point defect scattering [97], and phonon-dislocation scattering [44]. Regarding to the lattice thermal conductivity of superconductors, Klemens and Tewordt [44] studied the reduction of lattice thermal conductivity due to point defects.

The relaxation time is related to the phonon mean free path  $\Lambda_p$  as

$$\tau = \frac{\Lambda_p}{v_g} \tag{2.16}$$

where  $v_g$  is the phonon group velocity. In the case of phonon-electron scattering, the relaxation time can be calculated as the phonon mean free path, which is the inverse of attenuation coefficient  $\beta$  according to Pippard's theory [70].

$$\tau = \frac{\Lambda_p}{v_g} = \frac{1}{v_g \beta} \tag{2.17}$$

At liquid He temperatures (T < 4 K), the lattice thermal conductivity  $k_g$  is limited by phononelectron scattering and phonon-boundary scattering in defect free single crystals [44, 45]. According to Klemens [98], the electronic thermal conductivity is limited by electron-phonon scattering and electron-impurity scattering.

BCS theory [39] is the first microscopic theory of superconductivity which describes superconductivity as a microscopic effect caused by the condensation of Cooper pairs [99], for which they won the 1972 Nobel Prize. BRT [42] extended this theory to the thermal conductivity of superconductors by treating the excited states as quasi-particles to apply to the BTE. Tewordt [100] investigated the intrinsic electronic thermal conductivity of superconductors by introducing a nonequilibrium part of the distribution function into the corresponding BTE. He found the ratio of thermal conductivity in the superconducting state to that in the normal conducting state increases monotonically with  $T/T_c$  with a limiting slope close to 1.62 at  $T_c$ . Tewordt and Wölkhausen [80] established the theory of lattice thermal conductivity of high  $T_c$  superconductors by considering phonon-boundary scattering, phonon-point defect scattering, phonon-strain field scattering , and phonon-electron scattering, based on the BCS theory. The expression is shown in the following.

$$k_g(t) = A_1 t^3 \int_0^\infty dx \frac{x^4 e^x}{(e^x - 1)^2} \times [1 + A_2 t^4 x^4 + A_3 t^4 x^2 + A_4 t x + A_5 t x g(x, y)]^{-1}$$
(2.18)

where  $t = T/T_c$  is the reduced temperature,  $x = \frac{\hbar\omega}{k_BT}$  is the reduced frequency,  $A_1$  accounts for the scattering of phonons by boundary,  $A_2$  is for the scattering of phonons by point defect,  $A_3$  represents the scattering of phonons by strain field of sheetlike faults,  $A_4$  is for the scattering of phonons by strain field of dislocations, and  $A_5$  accounts for the scattering of phonons by electrons.

Connolly and Mendelssohn [53] measured the thermal conductivity of Nb and Ta below 1 K and analyzed their data into its electronic and lattice components. Their analysis agrees well with BCS theory. Anderson *et al.* [23] analyzed their thermal conductivity data of Nb by using Kinetic theory and separating the phonon mean free path into phonon boundary scattering and phonon dislocation scattering before and after deformation. Anderson and O'Hara [101] investigated the interaction of phonons by electrons to contribute to the thermal conductivity of Nb, they concluded that the mean free paths of transverse and longitudinal phonons are similar, which is close to  $4 \times 10^{-5} T^{-1}$  (cm K), in the normal state. Amrit [81] developed a model to investigate the effect of average grain size, Kapitza resistance at grain boundaries and number of grains on the thermal conductivity of poly-crystal Nb. The model predicted a strong spatial anisotropy in determining the thermal conductivity for more than 10 grains.

Koechlin and Bonin [41] modeled the thermal conductivity of superconducting Nb by parameterizing the BRT [42] expression according to Bardeen-Cooper-Schrieffer (BCS) theory [39] as

$$k = k_e + k_g = R(y) \left[\frac{\rho}{LT} + AT^2\right]^{-1} + \left[\frac{1}{D\exp(y)T^2} + \frac{1}{B\Lambda T^3}\right]^{-1}$$
(2.19)

where the two terms in  $k_e$  are due to electron-impurity scattering and electron-phonon scattering, respectively, and the two terms in  $k_g$  are due to phonon-electron scattering and phonon-boundary scattering, respectively. The term R(y) quantifies the condensation of normal conducting electrons into Cooper pairs [42].

Chandrasekaran *et al.* [84] improved this model by estimating directly from the temperature and heat flux measurements instead of estimating from the intermediate thermal conductivity results. This new estimating method groups coefficient of  $T^n$  in the model as different  $\beta$ s in the following

$$\beta_1 = \frac{\rho_{295K}}{LRRR}, \quad \beta_2 = a, \quad \beta_3 = \frac{1}{D}, \quad \beta_4 = \frac{1}{B\Lambda}, \quad \beta_5 = \frac{\Delta(T)}{k_B T_c}$$
 (2.20)

Equation 2.19 can be redefined as

$$k = R(y) \left[\frac{\beta_1}{T} + \beta_2 T^2\right]^{-1} + \left[\frac{\beta_3}{\exp(y)T^2} + \frac{\beta_4}{T^3}\right]^{-1}$$
(2.21)

with

$$y = \beta_5 \frac{T_c}{T} \tag{2.22}$$

Xu *et al.* [43] further improved this model by including the phonon dislocation scattering separately from boundary scattering in examining the lattice thermal conductivity. The model can be expressed as

$$k = R(y) \left[\frac{\rho}{LT} + AT^2\right]^{-1} + \left[\frac{1}{D\exp(y)T^2} + \frac{1}{B\Lambda T^3} + W_d\right]^{-1}$$
(2.23)

where  $W_d = KN_d/T^2$  is the thermal resistance of phonon dislocation scattering expressed in Eq. 2.4. Based on the properties of Nb, Klemens expressed  $W_d$  [44] for randomly distributed dislocations as

$$\frac{W_d T^2}{N_d} = 3.05 \times 10^{-15} \text{ m}^3 \text{K}^3 / \text{W}$$
(2.24)

According to Wasserbäch [4], Bross analyzed  $W_d$  in Cu for edge and screw dislocations. Bross obtained results to be 1.67 and 1.26 greater for edge and screw dislocations, respectively, than the values evaluated by Klemens of Eq. 2.8 [44]. Then the following equations are assumed for modeling the effect of dislocations to Nb.

$$\left(\frac{W_d T^2}{N_d}\right)^{\text{edge}} = 5.08 \times 10^{-15} \text{ m}^3 \text{K}^3/\text{W}$$
(2.25)

$$\left(\frac{W_d T^2}{N_d}\right)^{\text{screw}} = 3.85 \times 10^{-15} \text{ m}^3 \text{K}^3/\text{W}$$
(2.26)

Comparisons of data from [4] with fitting of the model are shown in Fig. 3.6 for a sample with RRR=250 after uniaxial straining of 14.7%. Fitting including the dislocation term improves comparison qualitatively and quantitatively with the experimental results [4], especially for samples after deformation and at low temperature. This improved agreement suggests that the observation by Koechlin and Bonin of a deviation in the fitting of their low temperature data using Eq. 2.19



Figure 2.7: Comparison between fitting with or without the dislocation term for deformed sample [4]. The estimated value of dislocation density is  $N_d = 3.83 \times 10^{14} \text{ m}^{-2}$ .

[41] may have been due to the lack of a phonon-dislocation scattering term. It also supports the idea that phonon-dislocation scattering contributes significantly to the thermal conductivity of Nb at low temperatures.

In Pippard's theory of free electrons in an isotropic metal, the longitudinal waves interact with the conduction electrons due to the density variation of the lattice contributing to the electric field of the space charge [102]. The transverse waves cause no density variation, therefore, no space charge. The attenuation coefficients for longitudinal and transverse waves can be written as

$$\beta_j = \frac{Nmv_F}{D_m v_j \Lambda_e F_j(z)} \tag{2.27}$$

where N is the number of conduction electrons per unit volume, m is the electronic mass,  $v_F$ is the Fermi velocity,  $D_m$  is the mass density,  $\Lambda_e$  is the electron mean free path, and  $F_i(z)$  is a

$T\Lambda_e (cm K)$	$I_1(z)$	$I_2(z)$
$10^{-8}$	2471	1493
$10^{-7}$	247.3	150.9
$7 \times 10^{-7}$	37.34	30.99
$9 \times 10^{-7}$	29.92	28.41
$10^{-6}$	27.36	27.77
$1.2 \times 10^{-6}$	23.58	27.25
$1.5 \times 10^{-6}$	19.89	27.59
$2 \times 10^{-6}$	16.30	29.57
$3 \times 10^{-6}$	12.90	35.58
$4.5 \times 10^{-6}$	10.78	46.20
$6 \times 10^{-6}$	9.786	57.44
$10^{-5}$	8.666	88.23
$10^{-4}$	7.336	79.38

Table 2.2: Numerical integration results for  $I_1(z)$  and  $I_2(z)$  [1]

function of phonon wave vector q and  $\Lambda_e$  ( $z = q\Lambda_e$ ), j denotes the longitudinal (l) or transverse (t) phonon branch.

For the longitudinal branch

$$F_l(z) = \frac{z^2 \arctan(z)}{3(z - \arctan(z))}$$
(2.28)

For the transverse branch

$$F_t(z) = 1 - \frac{3}{2z^2}((y^2 + 1)\frac{\arctan(z)}{z} - 1)$$
(2.29)

With these attenuation coefficients, the lattice thermal conductivity of metals considering phonon scattering with electrons only is obtained from Eq. 6.1

$$k_{pe} = \frac{D_m k_B^3 T^2}{(\pi^3 N m v_F \hbar^2) * I_j(z)}$$
(2.30)

where

$$I_j(z) = \frac{\pi}{6} \int_0^\infty \frac{zF_j(z)x^3 \exp(x)}{(\exp(x) - 1)^2} dx$$
(2.31)

where  $x = \frac{\hbar\omega}{k_BT}$  is the reduced frequency. The integrals  $I_j(z)$  have been tabulated by Lindenfeld and Pennebaker [103] in Table 2.2.

The number of free electrons decrease exponentially with temperature when it comes into superconducting state, because of the condensation of electrons into Cooper pairs. Assuming the number of free electrons at the normal conducting state is  $N_0$ , the expression for the number of free electrons at superconducting state is

$$N = N_0 \exp(-y) \tag{2.32}$$

Equation 2.30 becomes

$$k_{pe} = \frac{D_m k_B^3}{(\pi^3 N_0 m v_F \hbar^2) \times I_j(z)} \exp(y) T^2$$
(2.33)

The front part of the above equation  $\frac{D_m k_B^3}{(\pi^3 N_0 m v_F \hbar^2) \times I_j(z)}$  corresponds to the parameter *D* in Koechlin and Bonin's model [41] for phonon-electron scattering. The value of this parameter is calculated to be from  $6.85 \times 10^{-3}$  to  $7.55 \times 10^{-3}$  WK<sup>3</sup>m<sup>-1</sup> in the superconducting temperature range. This value is within 10% of the value D obtained from fitting with experimental data. It should be mentioned that there is no adjustable parameters in this model.

Table 2.3:	Research progr	ess on modelin	g $k$ of superc	onducting Nb
	1 0		0 1	e

Year	Principle Investigator	Contributions
1954	Wilson [37]	Theory of thermal conductivity of metals
1955	Klemens [44]	Phonon-impurity and phonon-dislocation scattering
1957	BCS [39]	Superconducting theory
1958	Klemens [38]	Scattering relaxation time in dielectrics
1959	Callaway [95]	Model of conduction in dielectrics
1959	BRT [42]	Theory of Superconductivity
1960	Klemens [97]	Phonon-point defect scattering
1962	Connolly [53]	Electronic and lattice contribution
1963	Holland [96]	Consider linear dispersion
1963	Tewordt [100]	Theory of electronic thermal conductivity
1964	Klemens and Tewordt [98]	Phonon-defect scattering for superconductors
		Cont'd on next page

Year	Principle Investigator	Contributions
1974	Anderson [23]	Mean free path of scatterings
1974	Anderson [101]	Longitudinal and Transverse phonons
1974	Kes [61]	Details of scattering relaxation time of Nb
1978	Wasserbach [4]	Phonon-dislocation scattering
1985	Wasserbach [3]	Phonon-electron scattering
1989	Tewordt [80]	Thermal conductivity of high $T_c$ superconductors
1993	Goodson [104]	Theory of $k$ of High $T_c$ superconductor
1996	Koechlin and Bonin [41]	Parameter estimation of thermal conductivity of Nb
2006	Amrit <i>et al.</i> [81]	Grains, Grain boundaries, Kapitza resistance
2010	Chandrasekaran et al. [84]	Estimation from heat flux and temperature
2017	Xu <i>et al</i> . [43]	Phonon-dislocation scattering explicitly

Table 2.3 – Cont'd from previous page

# 2.3 Conclusion

The thermal conductivities of superconducting Nb have been measured and modeled since the 1950s. Even so, there are some important omissions in our understanding of the relationships between k and other material properties including the effect of dislocations, impurities, and boundaries. This thesis is to explicitly examine these effects on the thermal conductivity of superconducting Nb and Ta.

#### **CHAPTER 3**

## EFFECT OF DEFORMATION ON THE THERMAL CONDUCTIVITY AND PHONON PEAK TEMPERATURE OF NB

## 3.1 Introduction

Manufacturing SRF cavities from Nb sheets requires large deformation that increases dislocation density [105], which has been shown to reduce the thermal conductivity of superconducting large grain Nb [3, 4, 8, 23, 57, 61]. Mendelssohn et al. [47] were the first researchers to attempt to quantify the phonon conduction in superconductors using the dislocation density  $N_d$  by measuring the thermal conductivity of superconducting Pb and Nb. A model from Van Bueren [48] was used to calculate the dislocation density for face-centered cubic (FCC) lattices (e.g., Pb), and the model of Klemens [44] was used to account for the temperature dependent thermal conductivity due to dislocations for body-centered cubic (BCC) materials (e.g., Nb). He found a local maximum thermal conductivity at 2.4 K for an unstrained Nb sample and the peak disappeared after 5% deformation. Wasserbäch measured the thermal conductivity of Nb after uniaxial straining of up to 22.2% [4]. An example of these data for a single specimen for which its conductivity was measured after each increment of deformation as replotted in Fig. 2.6. The thermal conductivity for temperatures colder than 3 K decreases with increasing deformation. Of particular note is the phonon peak in conductivity  $k_{pp}$  at approximately 2 K that decreases with increasing deformation. Wasserbäch examined the effect of deformation on conduction [3] by using a relaxation time approximation according to the Callaway model [95] with the relaxation time expression from Klemens [44]. Scattering of phonons by electrons, which is a significant factor at the working temperature of SRF cavities, was not included in the analysis. Chandrasekaran [8] measured the effect of deformation on k and quantified the role of subsequent heat treatment on the recovery of phonon peak and the decrease in dislocation density. A phonon peak that has disappeared after deformation can be partially restored with heat treatment of appropriate temperature and duration [85]. In analyzing



Figure 3.1: Measured phonon peak temperatures  $T_{pp}$  for undeformed, deformed, and annealed specimens as a function of the ratio of the thermal conductivity at the phonon peak  $k_{pp}$  to the local minimum thermal conductivity  $k_{lm}$ .

the effect of deformation on conductivity, Chandrasekaran used only  $k_{pp}$  to estimate the dislocation density. Koechlin and Bonin [41] used a simplified equation based on the BRT model [42] (see Eq. 2.19) to fit the experimental results. This equation was reparameterized by Chandrasekaran [84] for analysis and to reduce uncertainties in estimating the thermal conductivity of Nb. However, neither of these equations considered the scattering of phonons by dislocations, which might be the reason that Koechlin and Bonin noted discrepancies in fitting of k at temperatures lower than 2 K [41].

Figure 3.1 shows reported values of  $1.72 \le T_{pp} \le 2.35$  K for a number of studies of superconducting Nb [4, 7, 8, 12, 23, 61, 64, 78, 92] as a function of  $k_{pp}/k_{lm}$ , where  $k_{lm}$  is defined as the thermal conductivity at the local minimum, it typically appears at about T=3 K for Nb. Previous studies have indicated that  $k_{pp}$  is a function of dislocation density. This new observation suggests

that both  $k_{pp}$  and  $T_{pp}$  are functions of the dislocation density, and thus the deformation of Nb. This further suggests that the scattering of phonons by dislocations should be accounted for explicitly in the models of thermal conductivity. Figure 3.1 also shows the little discussed observation that the temperatures at which  $k_{pp}$  occurs increases as  $k_{pp}$  decreases. Typically, the temperature of the phonon peak  $T_{pp}$  of superconducting Nb is cited as occurring in the range  $1.8 \le T_{pp} \le 2$  K, but studies do not note a systematic variation. This addition of the dislocation term, and some of its implications, is considered here.

## 3.2 Analysis

The results in Fig. 3.1 represent measurements from several studies for as received, deformed, and annealed specimens. The temperature  $T_{pp}$  is plotted as a function of the ratio  $k_{pp}/k_{lm}$ . Chandrasekaran [8] showed that  $k_{pp}/k_3$ , where  $k_3$  is the thermal conductivity of 3 K, correlates inversely with dislocation density for a given specimen having undergone deformation or heat treatment. Although there is scatter in the data in Fig. 3.1, resulting from various sample processing histories and experimental techniques, there is a clear trend of  $T_{pp}$  decreasing with increasing value of  $k_{pp}/k_{lm}$ , and thus with decreasing dislocation density. Samples tested before deformation or after heat treatment have larger values of  $k_{pp}/k_{lm}$ . For T > 3 K, the effect of dislocation density can be neglected because the electron contribution dominates the thermal conductivity at this temperature range.

Figure 3.1 further shows that undeformed samples typically have colder  $T_{pp}$ , but greater  $k_{pp}$ . For deformation typically less than  $\epsilon = 3\%$  strain,  $k_{pp}$  decreases and  $T_{pp}$  increases. For uniaxial straining greater than 3%, the phonon peak disappears in most samples because of the increase of thermal resistance due to the scattering of phonons by dislocation. High purity samples, however, maintain a phonon peak after greater deformation (*e.g.*, the sample shown with RRR=1200 maintains a phonon peak after 4% uniaxial straining [4]). Deformation not only affects the value of thermal conductivity, but also has an apparent effect on  $T_{pp}$ . Heat treatment that causes dislocation annihilation (recovery) can reduce dislocation content [12] to partially or fully restore the phonon peak, depending on the annealed temperature and duration. Chandrasekaran [8] showed that 1000 °C for 4 hours is enough to nearly recover the phonon peak to that of the undeformed state.

As mentioned above, the scattering of phonons by dislocations should be included in the model, especially for Nb following deformation. Inspired by Klemens [44], a dislocation term may be added to Eq. 2.19 to account for the effect of dislocation as in Eq. 2.4.

## 3.2.1 Scaled sensitivity coefficient

To examine the effects of different scattering mechanisms, the scaled sensitivity coefficients of phonons scattered by boundaries, normal electrons, and dislocations are plotted with temperature.

## 3.2.1.1 Scaled sensitivity coefficient of boundary scattering

The scaled sensitivity coefficients are calculated as the product of the scattering parameters with the partial derivative of thermal conductivity over the parameters. Therefore, the scaled sensitivity coefficient has the same unit as the the thermal conductivity.

$$\chi_B = B \frac{\partial k}{\partial B} = \frac{1}{B\Lambda T^3} \left[ \frac{1}{D \exp(y)T^2} + \frac{1}{B\Lambda T^3} + \frac{KN_d}{T^2} \right]^{-2}$$
(3.1)

where  $\chi_B$  is the scaled sensitivity coefficient due to phonons scattered by boundaries.

## 3.2.1.2 Scaled sensitivity coefficient of normal electron scattering

Similarly, the scaled sensitivity coefficient of phonons scattered by electrons is shown as the following

$$\chi_D = D \frac{\partial k}{\partial D} = \frac{1}{D \exp(y)T^2} \left[ \frac{1}{D \exp(y)T^2} + \frac{1}{B\Lambda T^3} + \frac{KN_d}{T^2} \right]^{-2}$$
(3.2)

where  $\chi_D$  is the scaled sensitivity coefficient due to phonons scattered by electrons.



Figure 3.2: The scaled sensitivity coefficients of phonons scattered by boundary (parameter B) as a function of temperature. The B values vary from  $2 \times 10^3$  Wm<sup>-2</sup>K<sup>-4</sup> to  $10 \times 10^3$  Wm<sup>-2</sup>K<sup>-4</sup>.

### 3.2.1.3 Scaled sensitivity coefficient of dislocation scattering

The scaled sensitivity coefficient of phonons scattered by dislocations is shown as

$$\chi_N = N_d \frac{\partial k}{\partial D_d} = W_d \left[ \frac{1}{D \exp(y)T^2} + \frac{1}{B\Lambda T^3} + \frac{KN_d}{T^2} \right]^{-2}$$
(3.3)

where  $\chi_N$  is the scaled sensitivity coefficient due to phonons scattered by dislocations.

The scaled sensitivity coefficient of phonons scattered by boundaries is plot in Fig. 3.2. The values of D and  $N_d$  are kept the same and the B values vary from  $2 \times 10^3 \text{ Wm}^{-2}\text{K}^{-4}$  to  $10 \times 10^3 \text{ Wm}^{-2}\text{K}^{-4}$  to investigate the effect of phonons scattered by the physical boundaries. Similarly, for phonons scattered by normal electrons, the values of B and  $N_d$  are kept the same and the D values vary from  $2 \times 10^{-3} \text{ WK}^3\text{m}^{-1}$  to  $10 \times 10^{-3} \text{ WK}^3\text{m}^{-1}$ . The scaled sensitivity coefficient of phonons scattered by boundaries is plot in Fig. 3.3. For phonons scattered by dislocations, the



Figure 3.3: The scaled sensitivity coefficients of phonons scattered by normal electrons (parameter D) as a function of temperature. The D values vary from  $2 \times 10^{-3}$  WK<sup>3</sup>m<sup>-1</sup> to  $10 \times 10^{-3}$  WK<sup>3</sup>m<sup>-1</sup>.

values of B and D are kept the same and the  $N_d$  values vary from  $2 \times 10^{12}$  m<sup>-2</sup> to  $10 \times 10^{12}$  m<sup>-2</sup>. Taking the example of screw dislocations, the scaled sensitivity coefficient of phonons scattered by dislocations is plotted in Fig. 3.4.

Comparing Fig. 3.2 and Fig. 3.3 shows that  $\chi_B$  and  $\chi_D$  nearly have the same shape and magnitude, but of the maximums occurs at different temperatures. Figure 3.4 shows  $\chi_N$  is of different magnitude and sign. The different sign is because the lattice *k* decreases with  $N_d$ , while the lattice *k* increases with parameter B (*i.e.*, specularity) and D (*i.e.*, free electrons). It can be also seen from the figures of scaled sensitivity coefficients that the maximum occurs at about T=1.5 K for parameter B, higher than 2 K for parameter D, and  $T \approx 1.8$  K for parameter  $N_d$ . This observation confirms that boundary scattering dominates the lattice *k* at the lowest temperature, dislocation scattering affects the lattice *k* at about the phonon peak temperatures (*i.e.* 1.6 K < T < 2.2 K), and



Figure 3.4: The scaled sensitivity coefficients of phonons scattered by dislocations (parameter  $N_d$ ) as a function of temperature. The  $N_d$  values vary from  $2 \times 10^{12} m^{-2}$  to  $10 \times 10^{12} m^{-2}$ .

the phonons scattered by normal electrons start to affect the lattice k at T > 2 K.

# 3.3 Results and Discussion

Comparisons of data from [4] with fitting of Eq. 2.4 are shown in Fig. 3.5 for an undeformed sample with RRR=250 and Fig. 3.6 for the same sample after uniaxial straining of 14.7%. The differences in thermal conductivity between measurement data and the fitting results are also plotted for samples before and after deformation, to examine the accuracy of the model, as shown in Fig. 3.7 and Fig. 3.8. The residuals are similar in magnitude for the undeformed specimen in Fig. 3.7, because the thermal resistance due to phonons scattered by dislocations is negligible compared to that for phonons scattered by boundaries for samples before deformation. However, the residuals are significant in Fig. 3.8, but the fit that includes the dislocation term has residuals close to 0. The fit



Figure 3.5: Comparison between fit with or without dislocation term for an undeformed sample from [4]. The estimated value of dislocation density is  $N_d = 4.67 \times 10^{12} \text{ m}^{-2}$ . Both the fitted lines for temperatures lower than 1 K and higher than 2.5 K agree well with the experimental data. At 1.5 K < T < 2 K, fitting with the dislocation term has k values lower than the experimental results, while fitting without dislocation term has k values higher than the experimental results. The fit with or without the dislocation term provides similar accuracy for undeformed samples.

with the dislocation term included improves qualitatively and quantitatively with the experimental results [4], especially for samples after deformation and at low temperature. This improved agreement suggests that the observation by Koechlin and Bonin of a deviation in the fitting of their low temperature data [41] may have been due to the lack of a phonon-dislocation scattering term in their model. It also supports the idea that phonon-dislocation scattering contributes significantly to the thermal conductivity of Nb at low temperatures.

Several other sets of data from Wasserbäch [4] were fitted using the above method to examine the role of dislocation density and its effect on the thermal conductivity of Nb. These fits are shown



Figure 3.6: Comparison between fitting with or without dislocation term for deformed sample [4]. The estimated value of dislocation density is  $N_d = 3.83 \times 10^{14} \text{ m}^{-2}$ . Both the fitted lines for temperatures lower than 1 K and higher than 2.5 K agree well with the experimental data. At 1 K < T < 2.5 K, fitting with dislocation term matches well with the experimental results, while fitting without the dislocation term deviates from the experimental results. The fit with the dislocation term improves the accuracy for samples after deformations.

in Fig. 3.9 for screw dislocations and Fig. 4.8 for edge dislocations. Wasserbäch [4] calculated the dislocation density for sample K6 to be on the order of magnitude of  $10^{14}$  m<sup>-2</sup> after the last deformation (14.7%), without including phonon-electron scattering. The dislocation density extracted from the fit for the 14.7% deformation case is  $3.83 \times 10^{14}$  m<sup>-2</sup>. The dislocation density reaches  $10^{14}$  m<sup>-2</sup> after 10% deformation  $1.57 \times 10^{14}$  m<sup>-2</sup>. These are in reasonable agreement with values given by Wasserbäch [4], where he mentioned that the mean value of the dislocation density after transmission electron microscopy (TEM) measurement is in the range of 0.8 to  $1 \times 10^{14}$  m<sup>-2</sup>.

Using the enhanced model of Eq. 2.4, the effect of the magnitude of dislocation density on the



Figure 3.7: Differences between measurement data and the fit results of thermal conductivity with or without the dislocation term for an undeformed sample [4]. Here the differences, which are also called the residuals, are calculated as  $k_m - k_f$ , where  $k_m$  is the measured thermal conductivity and  $k_f$  is the fitted data.

thermal conductivity is studied. It is found that the phonons scattered by dislocations only affects the thermal conductivity when the dislocation density reaches a certain magnitude, *e.g.*,  $10^{12}$  m<sup>-2</sup>, shown in Fig. 3.11 by fitting the experimental results from [8] and varing the dislocation density in the model. The best fit is at  $N_d < 10^{12}$  m<sup>-2</sup>. However, the plots in Fig. 3.11 almost overlap with each other for  $N_d < 10^{12}$  m<sup>-2</sup>, indicating the scattering of phonons by dislocations is not important at these low values of dislocation density. The reason appears to be that phonon-boundary scattering dominates the heat conduction at low temperatures [46], and that the thermal resistance of phonons scattered by dislocations for  $N_d < 10^{12}$  m<sup>-2</sup> is much smaller than that of boundary scattering, as shown in Fig. 3.12. A threshold of dislocation density for contributing the thermal conductivity of



Figure 3.8: Differences between measurement data and the fit results of thermal conductivity with or without the dislocation term for a sample after 14.7% deformation [4]. Here the differences, which are also called the residuals, are calculated as  $k_m - k_f$ , where  $k_m$  is the measured thermal conductivity and  $k_f$  is the fitted data.

Nb is established here to be  $N_d = 10^{12} \text{ m}^{-2}$  using the proposed model.

The ratio of dislocation density after deformation  $N_d$  to that before deformation  $N_{d0}$  is plotted as a function of deformation (*i.e.*, uniaxial straining), shown in Fig. 3.13 for samples of varying RRR from [4]. This ratio increases with increasing deformation, the slope for samples with predominantly edge dislocations is larger than that for those with predominantly screw dislocations, implying that edge dislocations have greater influence on thermal conductivity. Note that the undeformed dislocation density is different for each sample, perhaps due to different RRR. For example, the best fit of the undeformed dislocation density for sample H2 (RRR=1200) is  $6.11 \times 10^{10}$  m<sup>-2</sup>, however, the values for the other three samples are approximately  $10^{12}$  m<sup>-2</sup> with similar RRR values for



Figure 3.9: Thermal conductivity of a sample from [4] (an example of predominantly screw dislocation) using curve fitting including the effect of dislocation. Here the dislocation density is estimated from Eq. 2.4 for the Cauchy strain  $\epsilon = 0\%$ , 3%, 7.3%, 10.3%, and 14.7%, respectively. Wasserbäch [4] provided a mean value of the dislocation density to be  $0.8 - 1 \times 10^{14}$  m<sup>-2</sup>, and the dislocation density measured at 14.7% is on the order of magnitude of  $10^{14}$  m<sup>-2</sup>.

185, 250, and 350.

# 3.4 Conclusion

For high performance of SRF applications, large values of thermal conductivity are desired for improved thermal stability. However, higher working temperature might yield significant savings in the total energy required for SRF cavity operations. Therefore, optimization of the phonon peak temperature and the value of thermal conductivity at that temperature becomes necessary for the thermal design of SRF cavities.



Figure 3.10: Thermal conductivity of a sample from [4] (an example of predominantly edge dislocation) using curve fitting including the effect of dislocation. Here the dislocation density is estimated from Eq. 2.4 for the Cauchy strain  $\epsilon = 0\%$ , 4%, and 8% respectively. Wasserbäch [4] provided a mean value of the dislocation density to be  $1 - 2 \times 10^{14}$  m<sup>-2</sup>, and the dislocation density measured at 8% is on the order of magnitude of  $10^{14}$  m<sup>-2</sup>.

Analysis of the thermal conductivity of superconducting Nb shows that in addition to  $k_{pp}$  decreasing after deformation, there is an increase in  $T_{pp}$ . The proposed model adds a phonondislocation scattering term that improves the accuracy of fits to experimental results, especially for samples after deformation. The proposed model can also be used to infer the dislocation density from measurements of k, as shown in Fig. 3.13. Results show that a threshold of dislocation density exists below which there is no significant effect on the thermal conductivity. A dislocation density smaller than  $10^{12}$  m<sup>-2</sup> has little contribution to the thermal conductivity of superconducting Nb, buy boundary scattering dominates at low temperatures, as shown in Fig. 3.11.



Figure 3.11: The effect of dislocation density on thermal conductivity of Nb at temperature between 1.5 K and 4.5 K. Equation 2.4 is used to fit the experimental results from [8]. The results are obtained by using the fitted parameters (*e.g.*, *B*, *D*, *a*) and changing  $N_d$ .



Figure 3.12: Comparison between phonon boundary thermal resistance and phonon dislocation resistance for different dislocation density by using the fitted parameters (*e.g.*, *B*, *D*, *a*) and changing  $N_d$ .



Figure 3.13: The ratio of dislocation density after deformation to that before deformation as a function of uniaxial straining, estimated by fitting the thermal conductivity from experimental results [4]. Samples H1, K6, H2, and K10 are deformed at temperatures of 77 K, 195 K, 295 K, and 1470 K, respectively. H1 and K6 had predominantly screw dislocations, H2 and K10 had predominantly edge dislocation.

#### **CHAPTER 4**

## MONTE CARLO MODEL OF THE LATTICE THERMAL CONDUCTIVITY OF NB

## 4.1 Introduction

The BTE has been successfully used to model phonon transport in both macro- and microscale heat conduction [95, 96, 106–108]. Solutions of the BTE are either through deterministic approaches [109–112] or stochastic methods (*i.e.*, Monte Carlo method) [9, 113, 114]. However, in the deterministic methods, the dispersion relationship has been omitted and the transverse and longitudinal polarization of the acoustic phonons has been replaced by a single effective polarization branch. The dispersion relation is important for modeling thermal transport because it provides details of the distribution of the properties of each phonon (*i.e.*, frequency, wave vector, and group velocity). In addition, in the deterministic methods, complex geometries result in high computational costs and the computational time increases exponentially with increasing dimension (i.e., from 2D to 3D). Moreover, nonlinear scattering events are also difficult to incorporate without a relaxation time approximation because the individual scattering events cannot be treated in isolation, where the relaxation time is defined as a time constant during which the distribution function relaxes to its equilibrium state. The stochastic Monte Carlo method is robust for modeling the dispersion of phonon transport from micro-scale to macro-scale. It is well suited for complex structures and multi-dimensions, and compared with molecular dynamics, it can reduce computational cost with large model size [115].

The Monte Carlo method has been widely used for phonon and electron transport [9, 113, 114, 116–128]. This method was first used to solve the BTE in modeling hot electrons in metal films [129]. Since then, it has been widely used in studying drift velocity and diffusion coefficients of charge transport in semiconductors [130], electron-electron interactions in GaAs [131], and thermal conductivity of thin films [114, 124], nanowires [118, 122], and nanotubes [116]. The Monte Carlo method has been successfully used to study ballistic phonon transport based on the

Debye model by assuming the same propagating speed and average lifetime interactions among phonons [113]. The temperature profile as a function of time evolution was predicted using these assumptions. Mazumder and Majumdar [114] presented a comprehensive solution of the BTE by considering phonon dispersion and polarization. Their simulation results agree well with the experimental results for Si at temperatures between 20 K and 300 K. Mittal and Mazumder [124] extended this model by including optical phonons, resulting in better matching with experimental observations for the thermal conductivity of Si thin-films. McGaughey and Jain [123] predicted the thermal conductivity of nanoscale Si thin films with arbitrary geometries. Lacroix et al. [9] developed a distribution function by considering the collision processes with respect to energy conservation during phonon scattering events in thin films. Their results provided good agreement with experimental results for phonon transport in both the ballistic and diffusion regimes. Chen et al. [118] modeled the phonon transport in single crystal Si nanowires by using a genetic algorithm including energy and momentum conservation in modeling the phonon-phonon normal and Umklapp scattering processes. Jeng *et al.* [120] modeled the thermal conductivity in nanoparticle composites of Si-Ge. They reproduced the earlier reported BTE results to validate their Monte Carlo model and then used the validated simulation method to study nanoparticle composites and showed that the thermal conductivity of nanoparticles can be smaller than that of the bulk value. Péraud et al. [126] proposed an energy-based variance-reduced Monte Carlo method. This method only needs to model the deviation in the energy of a particle population from its nearby equilibrium, with the equilibrium described analytically. This technique saves significant computational cost as compared with standard Monte Carlo methods.

## 4.2 Boltzmann Transport Equation

The general form of BTE for phonon can be written as [114]

$$\frac{\partial f}{\partial t} + v_g \cdot \Delta f = \left[\frac{\partial f}{\partial t}\right]_{scat} \tag{4.1}$$

where f(r, q, t) is the distribution function of an ensemble of phonons, which is related with position r, time t, and wave vector q, and  $v_g$  is the phonon group velocity. The left hand side of this equation represents the change of the distribution function due to motion or drift, and the right hand side represents the change due to collision or scattering. However, due to the nonlinear nature of scattering events, the relaxation time approximation is often used to simplify the scattering term by writing

$$\left[\frac{\partial f}{\partial t}\right]_{scat} = \frac{f - f_0}{\tau} \tag{4.2}$$

where  $\tau$  is the overall relaxation time due to various scattering mechanisms, such as phononphonon scattering, phonon-impurity scattering, phonon-boundary scattering, and phonon-electron scattering, and  $f_0$  is the phonon distribution function at equilibrium. Here phonon-impurity scattering refers to phonons scattered by impurities, phonon-boundary scattering refers to phonons scattered by physical boundaries, and phonon-electron scattering refers to phonons scattered by normal electrons.

## 4.3 Monte Carlo Solution Technique

The Monte Carlo method is used to solve the BTE under stochastic simulation and the relaxation time approximation. The energy-based variance-reduced technique [126] is used to model the lattice thermal conductivity of superconducting Nb by considering the phonon dispersion relation and the scattering of phonons by boundaries, normal electrons, and dislocations. The major steps for Monte Carlo simulation are outlined below.

## 4.3.1 Initialization

As the geometry of the studied material does not matter in the Monte Carlo simulation [9], a simple cubic cell stacks is chosen. The first step in the simulation, once medium, geometry, and mesh haven been chosen, is to initialize the state of an ensemble of phonons within each cell describing the material.

## 4.3.1.1 Number of particles

Using an integrated distribution function, the total vibrational energy of the crystal may be obtained as [9]

$$E = \sum_{p=LA,TA} \sum_{q} \left( \langle n_{q,p} \rangle + \frac{1}{2} \right) \hbar \omega$$
(4.3)

where  $\hbar$  is the reduced Planck constant,  $\omega$  is the phonon frequency,  $\langle n_{q,p} \rangle$  is the local thermodynamic phonon population with polarization p and wave vector q. The longitudinal and transverse acoustic branches are represented by LA and TA, respectively. The phonon population is assumed to follow the Bose-Einstein distribution function, that is

$$\langle n_{q,p} \rangle = \frac{1}{\exp(\frac{\hbar\omega}{k_B T}) - 1}$$
(4.4)

Introducing a density of state function of phonons,  $D_p$ ,

$$D_p(\omega)d\omega = \frac{Vq^2dq}{2\pi^2} \tag{4.5}$$

where V is the volume of the three-dimensional crystal. The group velocity of the phonons is defined based on the dispersion relation as

$$v_g = \frac{d\omega}{dq} \tag{4.6}$$

Then the total number of phonons in a given volume may be expressed as

$$N = V \sum_{p=LA,TA} \sum_{b=1}^{N_{bin}} \left[ \frac{1}{\exp(\frac{\hbar\omega_{b,p}}{k_B T}) - 1} \right] \frac{q_{b,p}^2}{2\pi^2 v_{gb,p}} g_p \Delta \omega$$
(4.7)

where  $g_p$  is the degeneracy of either the LA ( $g_p = 1$ ) or TA ( $g_p = 2$ ),  $N_{bin}$  is the number of spectral bins (chosen here to be 1000) in the range from 0 to  $\omega_{LA}^{max}$ , as in reference [114], and  $\Delta \omega = \omega_{LA}^{max}/1000$ .

Even for a small sample of material (*e.g.*, 10 nm silicon cube) at 300 K, there are approximately  $5 \times 10^5$  phonons [114]. Simulation can be reasonably accomplished only when the temperature is cold and the dimension size is small. Therefore, a weighting factor W is often used to save

computational cost by considering a bundle of phonons having the same properties. Then, the number of particles in the statistical computation  $N^*$  can be expressed as the actual number N divided by this weighting factor  $N^* = N/W$ . Péraud and Hadjiconstantinou [126] developed an efficient simulation method to model the deviation energy from the equilibrium by setting a hypothetical equilibrium temperature as the reference temperature and calculating the differences between the particle energy and corresponding equilibrium energy, making the computation time about 15 times faster to reach steady state. They also considered the energy-based method to provide a convenient and rigorous way to conserve energy automatically. In this method, each computational particle has the same amount of energy. The number of particles is defined as

$$N = V \sum_{p=LA,TA} \sum_{i=1}^{N_{bin}} \hbar \omega \left[ \frac{1}{\exp(\frac{\hbar\omega}{k_B T}) - 1} - \frac{1}{\exp(\frac{\hbar\omega}{k_B T eq}) - 1} \right] \frac{q^2}{2\pi^2 v_g} g_p s(i) \Delta \omega$$
(4.8)

where  $T_{eq}$  is the chosen reference temperature, s(i) is the sign of the particle, which is positive if  $T > T_{eq}$  and negative if  $T < T_{eq}$  to ensure that N is positive. Then, the energy is conserved by conserving the number of simulated particles.

During the initialization process, the first cell is set to the hot temperature  $T_h$  and the last cell to the cold temperature  $T_c$ , while all the other cells are also initially set to the cold temperature. The energy associated with each cell equals the number of particles in each cell as calculated by Eq. 4.8 and should match the calculated energy such that

$$N = \sum_{c=1}^{N_{cell}} \sum_{i=1}^{N^*} W * s(i)$$
(4.9)

#### 4.3.1.2 Frequency

The number of particles in the  $i^{th}$  spectral bin per unit volume is defined as

$$N_i(T) = \langle n(\omega_{0,i}, LA) \langle D(\omega_{0,i}, LA) \Delta \omega + 2 \langle n(\omega_{0,i}, TA) \langle D(\omega_{0,i}, TA) \Delta \omega$$
(4.10)

where the equilibrium distribution is evaluated at the central frequency of the spectral bin.

Once the number of particles is initialized, a frequency is assigned to each particle by a normalized number density function FD. According to Mazumder and Majumdar [114], this

function is written using the cumulative summation of the number of particles  $N_i(T)$  from Eq. 4.10, that is  $\Sigma_i^i = N_i(T)$ 

$$FD_{i}(T) = \frac{\sum_{j=1}^{l} N_{j}(T)}{\sum_{j=1}^{N_{bin}} N_{j}(T)}$$
(4.11)

Then, a random number  $R_1$  is selected from 0 to 1 and compared with the value of  $FD_i(T)$  in each bin. A bisection algorithm [114] is used to choose the  $i^{\text{th}}$  bin to satisfy  $FD_{i-1} \leq R_1 \leq FD_i$ . The actual frequency of the phonon is randomly chosen from another random number  $R_2$  (0 <  $R_2$  < 1) so that the frequency is linearly distributed between  $\omega_0 - \Delta \omega/2$  and  $\omega_0 + \Delta \omega/2$ , that is

$$\omega_i = \omega_{0i} + (2R_2 - 1)\frac{\Delta\omega}{2}$$
(4.12)

where  $\omega_{0i}$  is the central frequency of the *i*<sup>th</sup> spectral bin.

### 4.3.1.3 Polarization

The polarization (either LA or TA) is found from the ratio of the number of LA particles to the number of TA particles using Eq. 4.8 and the dispersion relation. The associated probability is expressed as

$$P_{LA}(\omega_i) = \frac{N_{LA}(\omega_i)}{N_{LA}(\omega_i) + N_{TA}(\omega_i)}$$
(4.13)

where  $N(\omega_i)$  is the number of particles in the branch of LA or TA with respect to the frequency.

Then, a third random number  $R_3$  is selected from 0 to 1. If  $R_3 < P_{LA}(\omega_i)$ , the phonon is on the LA branch, otherwise, it belongs to the TA branch.

### 4.3.1.4 Direction

Again, using the dispersion relation, the phonon group velocity can be determined. Assuming directional isotropy, the spherical coordinates, including the two angles  $\theta$  and  $\phi$ , can be chosen so that the corresponding directions are uniformly distributed in the  $4\pi$  full-space solid angle. The

direction  $\Omega$  can be determined using two random numbers  $R_4$  and  $R_5$  from 0 to 1, such that

$$\Omega = \begin{cases} \sin\theta\cos\phi\\ \sin\theta\sin\phi\\ \cos\theta \end{cases}$$
(4.14)

where  $\cos \theta = 2R_4 - 1$ , and  $\phi = 2\pi R_5$ 

### 4.3.1.5 Location

The last part of the initialization is to set a random location of each particle in each cell. Using the same method as before, the location of the  $n^{\text{th}}$  particle in cell u is expressed as

$$r_{n,u} = r_u + Ld_x R_6 i + Ld_y R_7 j + Ld_z R_8 k$$
(4.15)

where  $r_u$  are the coordinates of the cell u, *i*, *j*, *k* are coordinate directions,  $R_6$ ,  $R_7$ ,  $R_8$  are random numbers, and  $Ld_i$  is the size of the cube at each coordinate.

### 4.3.2 Drift process

After initialization comes the drift process, during which each particle will move according to its group velocity  $v_g$  and timestep  $\Delta t$ , such that  $r_{new} = r_{old} + v_g \Delta t$ . The timestep here is chosen so that  $\Delta t < Lc_x/v_g^{max}$  to consider all scattering events, and to avoid ballistic jumps over the cells,  $v_g^{max}$  is the maximum group velocity. The first and last cells are kept at constant hot and cold temperatures, and each particle coming into these two cells will be absorbed and re-sampled in order to keep the boundary cell acting as black-bodies. The rest of the medium is divided into several cells in the *x*-direction with the boundaries in the *y*- and *z*-directions being assumed to be adiabatic. When a particle reaches one of these adiabatic boundaries, it will be reflected either specularly (*i.e.*, angle of incidence equal to angle of reflection) or diffusively (*i.e.*, a random angle of reflection) according to the degree of surface specularities p ( $0 \le p \le 1$ , p = 0 purely specular, p = 1 purely diffuse). Thus, a random number  $R_9$  is drawn, when  $R_9 < p$ , a new particle drift direction is assigned using

Eq. 4.14, otherwise, the particle reflects specularly. After the drift process, the energy at each cell is calculated by the summation of the number of particles, and the temperature of each cell is computed by numerical inversion of Eq. 4.8.

## 4.3.3 Scattering

Phonons move inside a crystal structure with group velocity  $v_g$  and engage with scattering due to boundary, lattice structure, impurities, electrons, and defects. In the Monte Carlo simulation, the phonon boundary scattering process is carried out during the drift process, and the other scattering processes are treated independently from the drift. As phonons are mainly scattered by boundaries, electrons, and dislocations in Nb at superconducting temperatures [43], the phononphonon scattering and impurity scattering are neglected in this paper [95].

## 4.3.3.1 Phonon-electron scattering

Phonon-electron scattering is modeled according to Kes [61] by considering the condensation of electrons into Cooper pairs. Thus, the relaxation time for the phonon-electron scattering is computed as

$$\tau_{pe}^{-1} = g(x, T) E x T \tag{4.16}$$

where g(x, t) quantifies the ratio of the normal-conducting relaxation time to superconducting relaxation time and is a monotonically decreasing function of the non-dimensional frequency x( $x = \hbar w/(k_B T)$ ), T is the temperature and E is a fitting parameter found to be in the range of  $1.4 - 3 \times 10^9 \text{ K}^{-1} \text{s}^{-1}$ .

Assuming  $g(x,T) = g(0,T)(1 - x/(8x_m))$ , where  $g(0,T) = 2(\exp(y) - 1)^{-1}$  [61],  $x_m$  is the value of x when  $x^4 \exp(x)(\exp(x) - 1)^2$  has a maximum. Here  $x_m$  is calculated as 3.83. Then, the relaxation rate of phonon-electron scattering is expressed as

$$\tau_{pe}^{-1} = 2(\exp(y) - 1)^{-1} \left( 1 - \frac{x}{8 \times 3.83} \right) ExT$$
(4.17)

The probability of phonon scattered by electrons is

$$P_{pe} = 1 - \exp\left(-\frac{\Delta t}{\tau_{pe}}\right) \tag{4.18}$$

where  $\Delta t$  is the time step of Monte Carlo simulation.

A random number  $R_{10}$  is sampled to compare with the scattering probability  $P_{pe}$ . If  $R_{10} < P_{pe}$ , the phonon will be scattered by electrons. As the phonon-electron scattering is considered to be inelastic scattering, the phonon will be re-sampled after scattering, thus the frequency, wave vector, and velocity will be reassigned based on the dispersion relation and the number density function.

#### 4.3.3.2 Phonon-dislocation scattering

Phonon-dislocation scattering plays an important role in contributing to the thermal conductivity of superconducting Nb, especially after deformation [43]. The expression of the phonon-dislocation scattering relaxation rate is

$$\tau_{pd}^{-1} = A_0 \gamma^2 b^2 N_d w \tag{4.19}$$

where  $\gamma$  is the Grüneison constant ( $\gamma = 1.4$  for Nb [61]), *b* is the Burgers vector, and  $A_0$  is a constant. By applying the relaxation time of phonon-dislocation scattering into the Callaway model [95] and comparing the expression of the resistance in the Klemens model [44],  $A_0$  is evaluated to be 1.1398 for edge dislocations and 0.8639 for screw dislocations. Screw dislocations usually appear after deformation at temperatures lower than room temparure, whereas edge dislocations appear following high temperature deformation [4].

The probability of phonons scattered by dislocations is

$$P_{pd} = 1 - \exp(-\frac{dt}{\tau_{pd}}) \tag{4.20}$$

Similar to phonon-electron scattering, a random number  $R_{11}$  is assigned to compare with the probability  $P_{pe}$ . If  $R_{11} < P_{pd}$ , the phonon will be scattered by dislocations. Here, the phonon-dislocation scattering is considered to be elastic scattering, so that energy will be conserved after

scattering. Therefore, only the direction of velocity will be randomly assigned, with the frequency, wave vector, and magnitude of velocity remaining unchanged.

#### **4.3.4** Calculate the thermal conductivity of Nb

In the present study, the thermal conductivity is calculated knowing the heat flux of phonons through the medium for a given temperature gradient by directly applying Fourier's law. The temperature difference is preset so as to determine average thermal conductivity. The phonon heat flux is calculated along the temperature gradient as

$$q'' = \sum_{i=1}^{N} Ws(i) v_{gx}$$
(4.21)

where  $v_{gx}$  is the group velocity in x coordinate.

## 4.4 **Results and Discussion**

#### **4.4.1** Temperature profile at the diffusion limit

Comparison of the temperature profile at the diffusion limit was performed in a one-dimensional Si thin film with the boundary temperatures set as  $T_h = 310$  K and  $T_l = 290$  K. The equilibrium temperature was chosen to be the mean of  $T_h$  and  $T_l$  as  $T_{eq} = 300$  K to reduce the computational cost. The medium was divided into 20 cells, with each cell being  $5 \times 10^{-7}$  m by  $5 \times 10^{-7}$  m by  $5 \times 10^{-8}$  m, so the total length in the heat flux direction is  $20 \times 5 \times 10^{-8} = 1 \times 10^{-6}$  m = 1  $\mu$ m. The time step was chosen to be  $\Delta t = 1$  ps and there were 1000 spectral bins for sampling the frequency. The weighting factor was taken as  $W = 0.5 \times 10^{-16}$ . First, the diffusion limit ( $\Lambda << Lc$ ) was calculated at steady state. At the diffusion limit, phonon transport follows Fourier's law and the temperature profile is linear at steady state, as shown in Fig. 4.1. It shows that the simulation method successfully models the diffusion limit. One may notice that the simulated temperature is slightly lower than the diffusion temperature at T < 300 K and slightly greater than the diffusion temperature at T > 300 K. This is the result of the thermal conductivity of Si being temperature



Figure 4.1: Comparison of the temperature profile obtained using the Monte Carlo model with the usual Fourier conduction results in the diffusion limit. Six Monte Carlo simulations have been carried out to demonstrate the repeatability of the model, and the scatter in the results are shown.

dependent, it changes slightly from  $T_l = 290$  K to  $T_h = 310$  K. This is unaccounted for in the simple diffusion model.

### 4.4.2 Temperature profile at the ballistic limit

Comparison at the ballistic limit was also performed for the one-dimensional Si thin film. To enable a simulation using the same geometry of the material, the boundary cell temperatures were set to be  $T_h = 20$  K and  $T_l = 10$  K, because at lower temperatures, the phonon mean free path of Si increases and becomes too long for the physical geometry of the model. Keeping the size of the film unchanged, at low enough temperatures, the phonon mean free path will be comparable to the thickness of the film, causing the temperature distribution to reach the ballistic


Figure 4.2: Comparison of the temperature profile by Monte Carlo method with Stefen-Boltzmann equation in the ballistic limit. Six simulations have been carried out to demonstrate the repeatability of the Monte Carlo model.

limit. The temperature at the ballistic limit was calculated using Stephen-Boltzmann equation as  $T = ((T_h^4 + T_l^4)/2)^{0.25} = ((20^4 + 10^4)/2)^{0.25} = 17.1 K$ . Figure 4.2 shows good agreement between the Stephen-Boltzmann equation and the Monte Carlo method. The maximum error between the two solutions is approximately 2 percent. At low temperatures, the equilibrium distribution of phonons is nonlinear [132]. The small error in the Monte Carlo solution is caused by the non-linear distribution being approximated by a linear function of  $N_{bin} = 1000$  segments.

#### 4.4.3 Thermal conductivity in bulk Si

Comparison of the thermal conductivity in bulk Si was conducted using a 2  $\mu$ m length Si rectangle with the other two sides chosen as 500 nm × 500 nm. Simulation results of the thermal conductivity



Figure 4.3: Comparison of the thermal conductivity of bulk Si between different methods. The solid line is Eq. 4.22, the circles come from Monte Carlo simulation by Lacroix *et al.* [9], and the stars are the current simulation results.

of Si, as shown in Fig. 4.3, are compared with the Monte Carlo simulation results of Lacroix *et al.* [9] and the analytic equation for bulk Si [9] as

$$k_{Si} = \frac{\exp(12.570)}{T^{1.326}} \qquad (200K \le T \le 600K) \tag{4.22}$$

### 4.4.4 Size effect on thermal conductivity of Si nanowires

Comparison of the thermal conductivity of Si nanowires is carried out for different diameters of the nanowires (37 nm, 56 nm, and 115 nm). Monte Carlo simulation results are compared with the experimental data from Li *et al.* [10], as shown in Fig. 4.4. There is good agreement at the size of 37 nm and 56 nm. However, there are some discrepancies at d=115 nm for temperatures higher than 100 K, due to the fact that the same weighting factor was used for different sizes.



Figure 4.4: Comparison of the thermal conductivity of Si nanowire between current simulations (solid lines) and the data (triangles) from Li *et al.* [10]. Six simulations have been carried out and the error bars were plotted for the Monte Carlo simulation results.

Sample	Size	RRR	Dislocation	Deformation T	Heat Treatment
LS 1	3.9mm	350	Screw	195 K	NA
LS 2	3.2mm	1200	Edge	295 K	NA
LS 3	2mm	224	NA	NA	$3hr@800^{\circ}C \& @140^{\circ}C$

Table 4.1: Parameters for the samples used in the simulation

## 4.4.5 Simulation of the thermal conductivity of Nb

As the ability to characterize the thermal conductivity has been successfully demonstrated, it is next applied to model the lattice thermal conductivity of superconducting Nb. Samples used in the model are listed in Table. 4.1

To model the thermal transport of Nb at low temperatures, where phonons dominate the thermal conductivity, the phonon dispersion relation of Nb is replotted from Jani *et al.* [11] in Fig. 4.5 for



Figure 4.5: Phonon dispersion relation of Nb in the [1 0 0] direction, replotted from reference [11], where *a* is the lattice parameter, which refers to the physical dimension of unit cell in a crystal lattice, for Nb,  $a = 3.3 \times 10^{-10}$  m.

their experimental work. The phonon dispersion characterizes the relationship between frequency  $\omega$ , wavevector q, and group velocity  $v_g$ . It is the key to accurately model phonon transport and needs to be considered especially for this non-linear phonon dispersion.

### 4.4.5.1 Simulation of the lattice thermal conductivity of undeformed Nb

According to BCS theory [39], lattice thermal conductivity of superconductors consists of phononboundary scattering and phonon-electron scattering, where the phonon-defect scattering (*i.e.*, phonon-dislocation scattering) is included in the phonon-boundary scattering as the form of  $W_d \approx T^{-3}$  [41]. Considering phonon-dislocation scattering as the form of  $W_d \approx T^{-2}$  [98] in an extra term [43] demonstrated better fitting with the experimental results. Thus the phonon-



Figure 4.6: Simulation results of lattice k of undeformed Nb or Nb after heat treatment. Data used from undeformed samples from reference [4] and the sample after heat treatment of reference [12].

dislocation scattering is included in the simulation to model the lattice thermal conductivity of Nb. Dislocation density is introduced by deformation and it increases with the amount of deformation [51]. For undeformed Nb, phonon-dislocation scattering can be neglected because of the low dislocation density. The overall relaxation time in undeformed Nb  $\tau_{scat}$  consists of phonon-boundary scattering and phonon-electron scattering.

$$\tau_{scat}^{-1} = \tau_{pb}^{-1} + \tau_{pe}^{-1} \tag{4.23}$$

Simulation results of the lattice thermal conductivity of undeformed Nb sample and heat-treated sample are shown in Fig. 4.6 and compared with the fitting results using Eq. 2.4 of the phonon contributions. Heat treatment helps restore the phonon peak and increase the thermal conductivity because of the recovery (annihilation) of dislocations during heat treatment [8], where the phonon peak is the point at the local maximum thermal conductivity, which usually appears at around

2 K for Nb [85]. Therefore, the phonon-dislocation scattering can be neglected for the samples after appropriate heat treatment [12]. Simulation results agree well with the fitting results over the temperature range of SRF cavities when including phonon-boundary scattering, phonon-electron scattering, but neglecting phonon-dislocation scattering.

#### 4.4.5.2 Simulation of the lattice thermal conductivity of deformed Nb

For as-received Nb samples or deformed samples, the dislocation density is typically large thus the phonon-dislocation scattering cannot be neglected because it contributes significantly to phonon scattering in superconducting Nb [43]. In general, there are two basic types of dislocations, the edge dislocations and the screw dislocations. The difference between edge and screw dislocation is that the edge dislocation occurs when an extra half-plane of atoms exists in the middle of the crystal lattice whereas the screw dislocation occurs when the planes of atoms in the crystal lattice trace a helical path around the dislocation line [132]. According to reference [4], edge dislocations affect the thermal conductivity approximately 1.5 times more than screw dislocations because of their scattering mechanisms with phonons (there is a well-defined geometrical energy minimum due to the disturbed lattice positions, and the dislocation can easily wiggle along the line length; screw dislocation distort the lattice to a smaller extent). The overall relaxation time is expressed as the sum of phonon-boundary scattering, phonon-electron scattering, and phonon-dislocation scattering.

$$\tau_{scat}^{-1} = \tau_{pb}^{-1} + \tau_{pe}^{-1} + \tau_{pd}^{-1} \tag{4.24}$$

where  $\tau_{pd}$  is the phonon-dislocation scattering relaxation time either predominantly by edge dislocations or by screw dislocations.

Simulation results of the lattice thermal conductivity of Nb LS 1, an example of screw dislocations, have been compared with the fitting results for a sample before deformation, after 3% deformation, 7.3% deformation, and 10.3% deformation. The parameters are extracted from fitting and are included in the relaxation times of the simulation to consider different scattering mecha-



Figure 4.7: Simulation results of lattice thermal conductivity of Nb LS 1 from [4], an example of screw dislocations, for several strains.

nisms separately. The simulation results match well with the fitting results, as shown in Fig. 4.7. The phonon peak occurs at around 1.8 K in the simulation for sample LS 1 before deformation and it shifts to about 1.9 K after 3% deformation. After 7.3% deformation, the peak is diffuse and moves to 2.2 K. There is nearly no phonon peak after 10.3% deformation for this sample. Lattice thermal conductivity at temperatures lower than the phonon peak temperature is dominated by phonon-boundary scattering and follows the  $T^3$  relation. At temperature higher than the phonon peak, phonon-dislocation scattering is starting taking over for sample after large amount of deformation. Phonon-electron scattering contributes to the lattice thermal conductivity above 2 K and is dominant above 3 K.

Simulation results of the lattice thermal conductivity of Nb LS 2, an example of edge dislocations, have been compared with the fitting results for a sample before deformation, after 4%



Figure 4.8: Simulation results of lattice thermal conductivity of Nb LS 2, from [4] an example of edge dislocations, for several strains.

deformation, and 8% deformation, as shown in Fig. 4.8 The simulation results match well with the fitting results of both undeformed and deformed Nb samples by including the effects of phonon-dislocation scattering. Similar to sample LS 1, the steep phonon peak of the simulation results of sample LS 2 occurs at about 1.7 K. It moves to be around 2.2 K after 4% deformation with smooth peak. The phonon peak is much more diffuse after 8% deformation.

# 4.5 Conclusion

Monte Carlo solutions of the BTE have been successfully performed to model the lattice thermal conductivity of superconducting Nb given the phonon dispersion relationship of Nb. The validity of the simulation code was first demonstrated at the diffusion and ballistic limits. It was further demonstrated by comparing the simulation results of the thermal conductivity of bulk and nanowire

Si with analytical and experimental results [9, 10]. Then, the simulation method, incorporating the Nb dispersion relation and phonon-boundary, phonon-electron, and phonon-dislocation scattering, was used to calculate the lattice thermal conductivity of Nb for samples before deformation, after deformation to several strains, and after appropriate heat treatment. The simulation results agree well with the results extracted from experiments [4, 12]. The results show that boundary scattering dominates for temperatures below 2 K, where the phonon mean free path is comparable to the size of the sample, and phonon-electron scattering is important above 2 K and dominates the conductivity above 3 K. The phonon peak appears at temperatures of approximately 1.8 K in undeformed samples and shifts to a higher temperature after a small amount of deformation. Phonon-dislocation scattering decreases the lattice thermal conductivity and a large amount of deformation destroys the phonon peak. Appropriate heat treatment may restore the phonon peak by reducing the dislocation density. This simulation method may be extended to predict the thermal conductivity of Nb thin films in the application of Nb cladding on Cu to provide guidance for designing future generations of SRF cavities. In addition, with appropriate dispersion relations, this modeling technique may be used to model the lattice thermal conductivity of other superconductors, such as Ta and Nb<sub>3</sub>Sn, where conductivity is determined by considering the phonon scattering mechanisms.

#### **CHAPTER 5**

## EFFECT OF IMPURITIES ON THE LATTICE THERMAL CONDUCTIVITY OF NB

Thermal conductivity due to the contribution of electrons in metals consists of two forms of resistances to its flow. The first is from the interaction of electrons with interstitial and substitutional impurities in the metal ( $W_{ei}$ ), and the second is from the momentum exchange of the electrons with the lattice ( $W_{ep}$ ). These resistances act in series and the total resistance is shown as

$$W_e = W_{ei} + W_{ep} \tag{5.1}$$

The Wiedeman-Franz law relates the impurity limited electronic thermal conductivity  $k_{ei}$  and the electrical conductivity  $\sigma$  of the metal to the temperature as

$$\frac{k_{ei}}{\sigma} = LT \tag{5.2}$$

where L is the Lorenz constant and the electric resistivity  $\rho = \sigma^{-1}$ 

# 5.1 Residual resistance ratio (RRR)

Large values of thermal conductivity are required for good performance of SRF cavities to transfer the dissipated RF power to the liquid helium bath and prevent hot spots [31]. For bulk Nb cavities, this demands high purity. Because the thermal conductivity is usually related to the residual resistance ratio, which is an empirical indicator of the impurity concentration [36], at 4.2 K as

$$k_{4.2K} = \frac{\text{RRR}}{4} \tag{5.3}$$

where the RRR is defined as

$$RRR = \frac{\rho_{295K}}{\rho_{4.2K}}$$
(5.4)

where  $\rho_{295K}$  and  $\rho_{4.2K}$  are the resistivity of Nb at room and liquid helium saturation temperature at standard atmospheric pressure, respectively [31].

Impurity	N	0	C	Н	Hf	Ti	Zr	W	Ta	Mo
$\frac{\partial \rho_i}{\partial C_i} \times 10^{10}$	5.2	4.5	4.3	0.8	1.4	1.4	0.6-1.4	0.4-1.1	0.25	0.21

Table 5.1: Contributions of different impurities to  $\frac{\partial \rho_i}{\partial C_i}$  [1]

It is generally recognized that the resistivity of metals at low temperature is described as

$$\rho(T) = \rho_{res} + \rho_{ph}(T) + \rho_m \tag{5.5}$$

where  $\rho_{res} = \rho_{imp} + \rho_{def}$  is the residual resistivity at T $\approx 0$  K, which consists of electronimpurity scattering and the scattering by lattice defects;  $\rho_{ph}(T)$  represents the temperature dependent electron-phonon scattering, and  $\rho_m$  is the resistivity term in a magnetic field. For recrystallized niobium, the  $\rho_{def}$  contribution is small. In the absence of a magnetic field, the resistivity at low temperature is simplified as:

$$\rho(T) = \rho_{imp} + \rho_{ph}(T) \tag{5.6}$$

where  $\rho_{ph}(T)$  is also usually referred as the temperature dependent ideal resistivity. The resistivity due to the impurity contribution is expressed as

$$\rho(imp) = \sum \frac{\partial \rho_i}{\partial C_i} C_i \tag{5.7}$$

where  $C_i$  is the impurity concentration in atomic ppm and  $\frac{\partial \rho_i}{\partial C_i}$  has units of ohm-cm/at.-ppm. Therefore, Eq. 5.4 is updated to the following equation as

$$RRR = \frac{\rho_{295K}}{\rho_{ph}(4.2K) + \frac{\partial \rho_i}{\partial C_i}C_i}$$
(5.8)

The impurity content is typically consists of interstitial impurity atoms (*e.g.* H, N, O, and C) and substitutional impurity atoms (also called metallic impurities in Nb), including Ta, Mo, Zr, W, Ti, and Hf. The contributions of different impurities to  $\frac{\partial \rho_i}{\partial C_i}$  are listed in Table 5.1.

The interstitial impurities affect the RRR more significantly than the substitutional impurities. Therefore, attention should be paid to the chemical processing of the cavities. For example, the etching process will bring H impurities to the surface and the N baking process will add N impurities to the cavity. Typical RRR values for various types of Nb are listed in Table 5.2.

Table 5.2: Typical RRR values of Nb in relation to processings [2], where Cavities and Samples refer to post purified Nb origins, Other refers to other preparations.



Figure 5.1: Typical thermal conductivity data [4, 7, 13] related with RRR and strain, thermal conductivity increases with RRR at 4.2 K, while at 2 K, there is no relation between RRR and thermal conductivity. The phonon peak usually exists for appropriately heat treated sample while strain may destroy the phonon peak. (This figure is a repeat of Fig. 2.5), included here for the reader's convenience.

The predominant impurities in Nb from commercial cavities and post-purified cavities are interstitial impurities (including H, N, O, and C) [2]. The substitutional impurities contribute to higher RRR cavities from post-purified cavities below the theoretical limit, when there is 100% pure Nb.

Below the superconducting transition temperatures, the number of free electrons decreases

exponentially because of the formation of Cooper pairs [39]. Therefore, the phonon contribution will dominate the thermal conductivity at low temperatures ( $T < 0.2T_c$ ) and the electronic contribution is not important so that the effect of impurities can be neglected. Typical values of thermal conductivity with different RRR, strain, and heat treatment are plotted in Fig. 5.1. Thermal conductivity at 4.2 K increases with the increase of RRR values because the impurity atoms able to scatter with the free electrons decreases with the increase of RRR and the relation RRR =  $4k_{4.2}$ is descriptive. However, at temperatures lower than 3 K, especially at 2 K where the phonon peak usually appears, there is no relation between the thermal conductivity and RRR. The reason is that at this temperature, the phonon dominated thermal conductivity consists of the resistances due to phonon-boundary scattering, phonon-electron scattering, and phonon-dislocation scattering [43]. Phonons usually do not scatter with impurities at the lowest temperature because the sizes of the impurities are much smaller than the phonon mean free path of Nb at superconducting temperatures.

Fabrication of SRF cavities require surface processing, which includes buffered chemical polishing (BCP) and electropolishing (EP), and plastic deformation. These treatments increase the interstitial impurity content including O, H, and N and also form oxides, hydrides, and nitrides. These impurities affect the surface resistance of the SRF cavity to decrease the SRF performance [5]. Appropriate heat treatment might help decrease the near-surface concentration of impurities by dilution and also decrease the dislocation density. The effect of different types of impurities is discussed in the following.

## 5.2 Effect of oxygen diffusion

During low temperature baking (100 °C to 200 °C) of Nb SRF cavities, the oxygen diffuses from the surface oxide or an oxygen-rich interface layer just below the oxide into Nb, causing a decrease in the BCS surface resistance [5]. The average diffusion length of oxygen in Nb ( $x_d$ ) is expressed as

$$x_d = \sqrt{2D_O \exp(-E/RT)t} \tag{5.9}$$

where  $D_O = 0.015 \text{ cm}^2 \text{s}^{-1}$  is the diffusion coefficient for oxygen in Nb [133], E=112890 Jmol<sup>-1</sup> is the activation energy,  $R=8.31 \text{ Jmol}^{-1}\text{K}^{-1}$  is the universal gas constant, and *t* is the diffusion time.

After baking at 145 °C for 45 hours with a surface atomic oxygen concentration as 0.33%, which is close to the solubility of oxygen in Nb at 145 °C [5], the diffusion depth of oxygen spreads over 150 nm and drops by a factor of two within 50 nm. This thickness is comparable to the magnetic penetration depth of Nb, which is around 30 nm.

# 5.3 Effect of hydrogen diffusion

During fabrication process of SRF cavities, the BCP or EP processes are regularly used to etch the inner surface of the cavity to make the surface smooth and remove surface irregularities [5]. Niobium-hydride precipitates typically form at 100-150 K when the amount of hydrogen dissolved in niobium is significant, *e.g.*, the acid gets hot (T > 20 °C) during etching or too much H arrives at the cavity surface during EP process. This Nb-H effect is often called as the H-related Q-disease [134].

Chandrasekaran measured the thermal conductivity of superconducting Nb infused with saturated hydrogen (41.2% atomic concentration) [8]. He found a 25% reduction in thermal conductivity at the phonon peak for the a Nb bi-crystal exposed to 300 °C for 1 h in a 75% hydrogen, 25% nitrogen atmosphere at 0.5 atm., and a 15% reduction at the phonon peak for the bi-crystal specimen under uncooled BCP etch condition [8]. Several niobium-hydrogen phases have been identified depending on the concentration of hydrogen in Nb and the temperature of the specimen based on the Nb-H phase diagram [135]. The the Nb-H system is in the  $\alpha'$  phase after the hydrogen infusion at 300 °C. It transformed from  $\alpha'$  to  $\alpha' + \beta'$  during cooling at about 115 °C, and to  $\alpha + \beta$  at around 85 °C. In the  $\alpha$  and  $\alpha'$  phases, the hydrogen atoms are disordered and behaves like a gas [136]. However, in the  $\beta$  phase, hydrogen is ordered with well defined spacing [136] and behaves like a solid. Therefore, the mobility of the hydrogen atoms in the  $\beta$  phase is about two orders of magnitude lower than that in the  $\alpha'$  phase [137, 138]. After an 800 °C heat treatment for 2h the phonon peak was restored for one of the samples (heat treated at 800 °C for 2 hours before

hydrogen infusion) because of degassing the hydrogen [139].

# 5.4 Effect of nitrogen diffusion

To reduce the cryogenic operating cost accelerators while providing reliable operation, it is critical to improve the SRF performance of cavities, and thus improve the quality factor [5]. Nitrogen doping is an efficient way to improve this quality factor. Grassellino *et al.* [140] investigated the effect of nitrogen and argon doping of niobium on SRF performance. They found that 800 °C and 1000 °C nitrogen doping leads to low values of the microwave surface resistance (about 1.5 n $\Omega$ ), and an improvement in the efficiency of the accelerating structures up to a factor of 3. The reason for this enhancement of SRF performance is the formation of niobium nitride phases leading to a higher critical temperature than Nb. Dhakal *et al.* [141] also investigated the effect of low temperature (120-140 °C) nitrogen baking on the SRF performance of cavities and found an improvement in the quality factor after annealing at 800 °C for 3 hours followed by baking at 120–140 °C in a low partial pressure of nitrogen. However, the temperature has to be controlled carefully because a larger reduction (about 35%) of the quench field was observed when the baking temperature was 160 °C.

The interaction of impurities with the lattice typically appears in semiconductors or dielectric materials. Klemens [44] did a systematic investigation of impurity scattering by the lattice and it is discussed briefly in this chapter.

## 5.5 Substitutional atoms of different mass

In the case of an isolated impurity, the relaxation time of phonons scattered by an impurity is expressed as

$$\tau^{-1} = \frac{a^3}{G} (\frac{\Delta M}{M})^2 \frac{\omega^4}{4\pi v^3}$$
(5.10)

where *a* is the lattice parameter, G is the number of atoms in the crystal,  $\Delta M$  is the differences of mass between impurity and the lattice, *M* is the mass of lattice, and *v* is the group velocity based on the phonon dispersion.

# 5.6 Point defects in the MC Model

In the case of point imperfection, the relaxation time is expressed as

$$\tau_{pi}^{-1} = \frac{3a^3}{G} S^2 \frac{\omega^4}{\pi v^3}$$
(5.11)

where  $S^2 = S_1^2 + S_2^2 + S_3^2$ .

$$S_1^2 = \frac{\Delta M}{M} \frac{1}{2\sqrt{3}} \quad S_2^2 = \frac{\delta v^2}{v^2} \frac{1}{\sqrt{6}} \quad S_3^2 = -Q\gamma \frac{\Delta R}{R} \sqrt{\frac{2}{3}}$$
(5.12)

where Q is a constant, R is the atomic radius, and  $\gamma$  is the Grüneisen constant.

Mazumder and Majumdar [114] simplified this relaxation time as the following:

$$\tau_{pi}^{-1} = \alpha \sigma \rho |V_g| \tag{5.13}$$

where  $\alpha$  is constant with the order of unity,  $\rho$  is the impurity concentration of point defect, and  $\sigma$  is the scattering cross-section expressed as

$$\sigma = \pi R^2 (\frac{\chi^4}{\chi^4 + 1})$$
(5.14)

where  $\chi = R|q|$ .

The effect of impurity concentration on the temperature profile of Si thin film is shown in Fig. 5.2 by the Monte Carlo simulation [114]. The specularity is set to be unity so that the boundary scattering is neglected in the simulation. It is shown from the figure that with increasing defect density, the temperature drop across the film increases.

# 5.7 Monte Carlo simulation considering impurity scattering

The effect of interstitial impurities (H and N) on the lattice thermal conductivity of superconducting Nb is investigated using the Monte Carlo simulation.

Simulation of the lattice thermal conductivity of Nb is shown in Fig. 5.3 for different levels of H concentration and in Fig. 5.4 for different levels of N concentration. The fitting results were conducted using data from reference [4] using the enhanced model [43] without considering



Figure 5.2: Temperature profile of Si for different levels of impurities from  $10^{23}$  m<sup>-3</sup> to  $10^{28}$  m<sup>-3</sup>. The temperature profile changes gradually from the ballistic limit for the lowest defect density  $10^{23}$  m<sup>-3</sup> to diffusion limit at the highest defect density  $10^{28}$  m<sup>-3</sup>.

impurity scattering. The simulation results with no impurity atoms are close to the fitting results. There are no significant differences between the simulated lattice thermal conductivity from phonon-impurity scattering until the concentration reaches a certain amount. To significantly affect the lattice thermal conductivity of Nb, the concentration has to reach  $10^{28}$  m<sup>-3</sup> and  $10^{27}$  m<sup>-3</sup> for H and N impurity atoms, respectively. In these cases, Nb is saturated with H or N, and the atomic ratio of the impurity to Nb lattice is close to 1.

Although k is correlated with RRR for  $T \gtrsim 0.3T_c$  (specifically k(4.2 K) = RRR/4), a few measurements [4, 8] suggest that  $k_{pp}$  may be correlated with RRR. See, for example, Fig. 2.5. This result is unexpected based on the BRT model of k of superconductors [42] and the theory of metals [37]. The extended model (Eq. 2.4) offers an explanation based on the results of Wasserbäch [4]



Figure 5.3: Simulation of the lattice thermal conductivity of Nb with different levels of H impurities compared with fitting results without H impurities. The lattice thermal conductivity values are similar for low concentrations of H impurities (*i.e.* No impurity,  $10^{26}$  m<sup>-3</sup>, and  $10^{28}$  m<sup>-3</sup>), but at the impurity concentration of  $10^{29}$  m<sup>-3</sup>, the lattice thermal conductivity decreased by 20% at the phonon peak.

and Chandrasekaran [8]. The coefficient of the phonon-boundary scattering term varies with RRR. In samples with large  $k_{pp}$ , the dislocations have little effect and the value of the  $k_{pp}$  is a result of the balance of the phonon-boundary and phonon-electron scattering.

Simulations of the lattice thermal conductivity of Nb were also conducted for two different samples from reference [8]. The first sample was heat treated at 800  $^{\circ}$ C for 2 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300  $^{\circ}$ C for 1 hour, and then heated again at 800  $^{\circ}$ C for 2 hours. The simulated lattice thermal conductivity without considering impurity atoms matches with the fitting results with no impurities, as shown



Figure 5.4: Simulation of the lattice thermal conductivity of Nb with different levels of N impurities compared with fitting results without N impurities. The lattice thermal conductivity values are similar for low concentrations of N impurities (*i.e.* No impurity,  $10^{26}$  m<sup>-3</sup>, and  $10^{27}$  m<sup>-3</sup>), but at the impurity concentration of  $10^{28}$  m<sup>-3</sup>, the lattice thermal conductivity decreased by 15% at the phonon peak.

in Fig. 5.5. After H infusion, the concentration of H is estimated to be around  $3.854 \times 10^{28} \text{m}^{-3}$ . The simulated lattice thermal conductivity agrees well with the data extracted from the experiment results, however, the H impurity concentration has to reach  $2.2 \times 10^{29} \text{m}^{-3}$  in the simulation, which is 6 times greater than the estimated concentration, to match with the data, shown in Fig. 5.5. The data extraction process is to obtain the lattice thermal conductivity of Nb considering impurity scattering from the difference between the total measured thermal conductivity and the electronic thermal conductivity, as shown in Eq. 5.15. Since the electronic thermal conductivity, which dominates at T > 3K, does not change with impurity content for the first sample above 3 K, the main influence of H impurity will be on the lattice thermal conductivity.



Figure 5.5: Lattice thermal conductivity of Nb by considering the effect of phonon impurity scattering. This sample was heat treated at 800 °C for 2 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300 °C for 1 hour, at last it was heat treated again at 800 °C for 2 hours. The last heat treatment to degas the H impurity recovers the thermal conductivity at phonon peak.

$$k_p = k - k_e \tag{5.15}$$

# 5.8 Phonon-impurity scattering

Imperfections typically affect the lattice thermal conductivity, however this phenomenon changes at the lowest temperatures [22]. The reason can be explained as follows. When the temperature decreases, the wavelength of the lattice increases to be much greater than the atomic spacing, *e.g.*, the dominant wavelengths reach around 100 times the atomic radius at 1 K. These long wavelengths



Figure 5.6: Lattice thermal conductivity of Nb by considering the effect of phonon impurity scattering. This sample was heat treated at 140 °C for 48 hours and 1100 °C for 4 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300 °C for 1 hour, at last it was heat treated again at 800 °C for 2 hours. The last heat treatment has no effect to degas the H impurity and does not recover the thermal conductivity at phonon peak.

are nearly unaffected by the imperfections on the order of the atomic scale, but are affected by the physical boundary of a crystal. However, the wavelengths of electrons depend little on the temperature and are on the same order as the atomic scale. Therefore, the imperfections will be the dominant scatter for electrons at low temperatures.

Moreover, at unrealistic high concentration of impurities, the phonons will be scattered by the imperfections and affect the lattice thermal conductivity. The expression of the relaxation time of phonon-impurity scattering  $\tau_{pi}$  is defined as

$$\tau_{pi}^{-1} = A_1 \omega^4 \tag{5.16}$$

where  $A_1$  is constant expressed as

$$A_1 = \frac{V\Gamma}{4\pi v_s^3} \tag{5.17}$$

where  $\Gamma = \sum_{i} f_{i} (\frac{\Delta M}{M})^{2}$ ,  $f_{i}$  is the atomic fraction of the impurity, M is the mass of the host atom and  $\Delta M$  is the mass differences between host atom and impurity, and V is the volume.

The second sample was heat treated at 1100 °C for 4 hours first, then it was infused with hydrogen in a 75% H2, 25% N2 environment of 0.5 atm pressure at 300 °C for 1 hour, at last it was heat treated again at 800 °C for 2 hours. The simulated lattice thermal conductivity without considering impurities matches well with the fitting results of no impurities, as shown in Fig. 5.6. After H infusion, the concentration of H impurity is estimated to be around  $3.9 \times 10^{28} \text{m}^{-3}$ . The simulated lattice thermal conductivity agrees well with the data extracted from the experiment results, and also with the fitting results of considering phonon-impurity scattering. In this sample, the H impurity concentration needs to reach  $8 \times 10^{28} \text{ m}^{-3}$  in the simulation, which is about two times greater than the estimated concentration, to match with the data extraction and fitting results, as shown in Fig. 5.6.

# 5.9 Effect of Grain Boundary scattering on the lattice thermal conductivity of superconducting Nb

A grain boundary is defined as the interface between two grains, or crystallites, in a polycrystalline material. Grain boundaries are considered as 2D defects in the crystal structure, and tend to decrease the electrical and thermal conductivity of the material [98].

# 5.9.1 Models

Nan and Birringer investigated the influence of grain size and grain–boundary thermal resistance (Kapitza resistance  $R_K$ ) on the effective thermal conductivity  $k_E$  of poly-crystals. They obtained

the normalized thermal conductivity  $(k_E/k_0)$  in terms of the Kapitza length  $L_K$  and the grain size d. For an isotropic poly-crystalline solid composed of spherical crystals of equal size, they showed

$$\frac{k_E}{k_0} = \left(1 + \frac{\phi L_K}{d}\right)^{-1} \tag{5.18}$$

where  $k_0$  is the bulk thermal conductivity of a single crystal,  $\phi$  is a constant depending on the aspect ratio of the crystallite ( $\phi \approx 2$ ). This equation is powerful in that it states that the normalized thermal conductivity ( $k_E/k_0$ ) must scale as a function of ( $L_K/d$ ). It also implies that the different microscopic scattering mechanisms take place in poly-crystals, that is, phonon scattering at grain–grain interfaces (via  $R_K$ ) and the phonon–phonon, phonon–boundary, and phonon–defect scattering.

Yang *et al.*[5] developed a simple heat transfer model to analyze their measurements of the thermal conductivity of nano-crystalline yttria-stabilized zirconia (YSZ). They investigated experimentally grain sizes ranging from 10 to 100 nm using the  $3\omega$  method at 300 K. In their model, they first considered the case when  $\lambda < \Lambda_{mfp} < d$  where  $\lambda$  and  $\Lambda_{mfp}$  are the phonon wavelength and mean free path, respectively. If  $\Delta T_0$  is the temperature jump over the bulk single crystal, then the heat flux is simply given by  $q'' = k_0 T_0/d$ . In reality, the single crystal has an effective thermal conductivity *k* due to grain boundary scattering, which gives rise to a grain– grain boundary thermal (Kapitza) resistance. This latter is related to the heat flux by the equation  $q'' = \Delta T_{gb}/R_K$ , where  $\Delta T_{gb}$  is the temperature jump at a grain–grain boundary. The following expression is obtained

$$\frac{k_E}{k_0} = (1 + \frac{L_K}{d} + \mu)^{-1}$$
(5.19)

where  $\mu = \Delta B / \Delta T 0$  is an artificially introduced parameter which accounts for the situation where  $\lambda > d$ ,  $\Delta T 0$  is the temperature jump across a single crystal and  $\Delta B$  is a temperature correction to  $\Delta T 0$ .

This expression is similar to Eq. 5.18 except for the aspect ratio  $\gamma$  and the artificially introduced parameter  $\mu$ . The latter accounts for the situation when  $\lambda \ge d$ . Yang *et al.*[5] fitted their data with

the above expression, using a unique value of  $R_K$  in the entire *d* range. The excellent agreement testifies that the reduction in the conductivity is due to decreasing grain sizes.

Amrit [81] modified Eq. 5.18 and Eq. 5.19 by considering an arrangement of single crystals to account for the spatial anisotropy of poly-crystals:

$$\frac{k_E}{k_0} = (1 + \frac{n_i - 1}{n_i} \frac{L_K}{d})^{-1}$$
(5.20)

where  $n_i$  is the number of grains in the direction *i*, and  $n_i$  adjacent grains have  $n_i - 1$  interfaces.

In this equation, the thermal conductivity not only depends on the Kapitza length  $(L_K)$  and grain size, but also is a function of the number of grains. He concluded that the thermal conductivity decreased strongly with the increase of the number of grains up to 10. He referred to this change of thermal conductivity as anisotropy [81]. When the number of grains exceed 10, the effect of the anisotropy of thermal conductivity disappeared.

# 5.10 Conclusion

Fabrication of SRF cavities requires surface processing (*i.e.*, BCP, EP, and plastic deformation) which increases the interstitial impurity content. These impurities affect the surface resistance of the SRF cavity to decrease the SRF performance [5]. RRR values are a function of different interstitial impurities and substantial impurities, and RRR is an indicator of the thermal conductivity at 4.2 K based on  $RRR = 4 \times k_{4.2K}$ . Because the impurity atoms typically scatter electrons for metals rather than phonons, it affects the electronic thermal conductivity, but not the lattice thermal conductivity, except for special conditions (*i.e.* the impurities are saturated in the Nb sample). Monte Carlo simulations of the effect of H impurity and N impurity atoms on the lattice thermal conductivity show that only at the highest concentrations of impurity (*i.e.*,  $10^{28}$ m<sup>-3</sup> for H and  $10^{27}$ m<sup>-3</sup> for N), the lattice thermal conductivity will decrease with increasing impurity concentration. This is similar to the definition of the threshold of dislocation density. Here the threshold of impurity concentration is calculated as  $10^{28}$ m<sup>-3</sup> and  $10^{27}$ m<sup>-3</sup> for H and N impurities, separately. Simulations of the lattice thermal conductivity of H atomic concentration at 41.2% match the the fitting results

with experimental data considering the effect of phonon-impurity scattering. However, there are still some discrepancy on the impurity concentration. The reason might be that the measured thermal conductivity above 3 K remained the same after saturation of H atoms, which does not agree with the BRT theory in the electronic dominated temperature range, where electrons are mainly scattered with impurities and the thermal conductivity will decrease with the increase of impurity concentration. Therefore the data extraction overestimate the effect of impurities on the lattice thermal conductivity. Various scattering relaxation time expressions for phonon-impurity scattering have been listed for point defect, substantial impurity at different mass, separately. The effect of grain boundary scattering is also studied and it will affect the lattice thermal conductivity for poly-crystal Nb samples because of the increasing number of grains than single crystal samples.

#### **CHAPTER 6**

# CALCULATION OF THE LATTICE THERMAL CONDUCTIVITY OF NB CONSIDERING PHONON DISPERSION

# 6.1 Introduction

Lattice thermal conductivity of superconducting Nb consists of phonon-boundary scattering and phonon-electron scattering for samples before deformation, without the effect of dislocations. A term considering phonon-dislocation scattering has been added in the model from Koechlin and Bonin [41] to account for the effect of deformation [43] on the lattice thermal conductivity of superconductors. This effect was studied in Chapter 3. Numerical simulation using Monte Carlo simulation of the lattice thermal conductivity of superconducting Nb is robust and takes account of individual scattering mechanisms as well as including the phonon dispersion. It is also convenient for complex structures. This model was studied in Chapter 4. However, this method is not a closed form solution to the BTE so will have some statistic errors. Investigation of the closed form solution of the BTE is critical to understand the mechanism in contributing to the lattice thermal conductivity of superconductors.

# 6.2 Model

The Callaway model [95] has been widely used to model the thermal conductivity of dielectric materials and semiconductors with the relaxation time approximation. The TW model [80] which considers the condensation of electrons into Cooper pairs is often used for high  $T_c$  superconductors. These two models are similar, and the general form is expressed as

$$k_g = \frac{k_B^3}{\hbar^3} \frac{k_B}{2\pi^2 v_g} T^3 \int_0^{\Theta_D/T} \frac{\tau \exp(x) x^4}{(\exp(x) - 1)^2} dx$$
(6.1)

where  $x = \frac{\hbar\omega}{k_BT}$  is the non-dimensional frequency,  $v_g$  is the group velocity, here taken as a constant with the value of sound velocity (no dispersion considered),  $\tau$  is the overall relaxation time of



Figure 6.1: Phonon dispersion of Nb at [1 0 0] direction, replotted from the experimental data of [14]

different scattering mechanisms, including phonon-phonon scattering, phonon-electron scattering, phonon-impurity scattering, phonon-boundary scattering (phonon-grain boundary scattering), and phonon-dislocation scattering, and  $\Theta_D$  is the Debye temperature, here taken as  $\Theta_D$ =275 K for Nb.

The model in Eq. 6.1 does not include phonon dispersion and assumes isotropy in the longitudinal and transverse acoustic directions. The following model includes phonon dispersion and treats the phonon ensemble separately to calculate the lattice thermal conductivity of superconducting Nb and is expressed as the following

$$k_g = \frac{1}{2\pi^2 k_B T^2} \int_0^{q_{max}} (\hbar\omega)^2 \omega^2 \frac{\tau \exp\left(\frac{\hbar\omega}{k_B T}\right)}{\left[\exp(\frac{\hbar\omega}{k_B T}) - 1\right]^2} dq$$
(6.2)

where  $\omega$  is the phonon frequency, q is the wave vector.

#### 6.2.1 Phonon dispersion of Nb

To account for the effect of phonon dispersion, two polarization branches have been included in the integration, namely longitudinal acoustic (LA) branch and transverse acoustic (TA) branch, where LA branch has one direction and TA branch has two directions. The experimental phonon dispersion of Nb is replotted from reference [14], as shown in Fig. 6.1. The optical direction is neglected in the expression because of its low group velocity to contribute to the lattice thermal conductivity [114]. For each wave vector q, the group velocity  $v_g$  is related to  $\omega$  following the relation of  $d\omega/dq = v_g$ . This allows consideration of variable phonon group velocities.

The relaxation times of different scattering mechanisms are expressed as the following to be inserted into the extended Callaway model of Eq. 6.2.

### 6.2.2 Phonon-boundary scattering

The phonon-boundary scattering accounts for the traveling of phonon particles when they hit the physical boundary of the specimen. This scattering is considered as elastic scattering, therefore the energy is conserved by the scattering event. The properties (frequency, wave vector, and the magnitude of group velocity) of the scattered phonons will remain the same. The phonons will reflect back either specularly or diffusively depending on the specularity of the surface. The relaxation time  $\tau_{pb}$  is expressed as

$$\tau_{pb}^{-1} = \frac{v(1+p)}{l(1-p)} \tag{6.3}$$

where *p* is the specularity of the surface which relates the scattering directionality to the surface roughness. If p = 0, it means purely diffusive reflection, p = 1 refers to purely specular reflection.

### 6.2.3 Phonon-electron scattering

Phonon-electron scattering occurs when the traveling phonons collide with the normal electrons (note that electrons which form in Cooper pairs do not scatter phonons). This scattering is considered as unharmonic collision process, which is also called the inelastic scattering. Therefore,

the energy is not conserved after the scattering, so the properties (frequency, wave vector, and group velocity) of the scattered phonons will be changed. In the Monte Carlo simulation, this process is carried out during the re-sampling process of the phonons using random numbers. A new phonon will be drawn in the corresponding temperature range to replace the old phonon with new properties. The expression of the relaxation time of phonon-electron scattering ( $\tau_{pe}$ ) is

$$\tau_{pe}^{-1} = g(x, T)ExT = 2(\exp(y) - 1)^{-1}(1 - \frac{x}{8 \times 3.83})ExT$$
(6.4)

where g(x, t) quantifies the ratio of the normal-conducting relaxation time to superconducting relaxation time and is a monotonically decreasing function of the non-dimensional frequency x ( $x = \hbar w/(k_B T)$ ), E is a fitting parameter that is found to be in the range of  $1.4 - 3 \times 10^9 \text{ K}^{-1} \text{s}^{-1}$  [61]. The constant 8 in the expression is obtained by comparing the integrated thermal conductivity due to phonon-electron scattering only with the thermal conductivity from the model of reference [41].

#### 6.2.4 Phonon-dislocation scattering

The phonon-dislocation scattering happens when the traveling phonons collide with the dislocations, either screw dislocations or edge dislocations. This scattering process is also considered as elastic scattering, therefore the energy is conserved by the scattering event. The properties (frequency, wave vector, and the magnitude of group velocity) of the scattered phonons will remain the same, while the direction of the group velocity is resampled using random numbers in the Monte Carlo simulation. The expression for the phonon-dislocation scattering relaxation time  $\tau_{pd}$  is

$$\tau_{pd}^{-1} = A_0 \gamma^2 b^2 N_d w \tag{6.5}$$

where  $\gamma$  is the Grüneison constant ( $\gamma = 1.4$  for Nb [61]), *b* is the Burgers vector, and  $A_0$  is a constant related to the dislocation mechanisms (*i.e.*, edge dislocations or screw dislocations). By applying the relaxation time of phonon-dislocation scattering to the Callaway model [95] and comparing the expression of the resistance in the Klemens model [44],  $A_0$  is calculated to be 1.1398 for edge dislocations and 0.8639 for screw dislocations. Screw dislocations usually appear after deformation at temperatures lower than room temperature, whereas edge dislocations appear following high temperature deformation [4].

#### 6.2.5 Total relaxation time

The total relaxation time of different scattering mechanisms studied above is obtained by using the Matthiessen's rule as

$$\tau^{-1} = \tau_{pb}^{-1} + \tau_{pe}^{-1} + \tau_{pd}^{-1} \tag{6.6}$$

This total relaxation time in Eq. 6.6 is inserted in the extended Callaway of Eq. 6.2 by considering the phonon dispersion. The lattice thermal conductivity  $k_g$  is calculated by integrating over the wave vector from 0 to the maximum values. However, this model is limited by the requirement that all of the relaxation times being considered to be of the same form as shown in Eq. 6.6.

# 6.3 **Results and Discussions**

Calculation of lattice thermal conductivity of superconducting Nb was performed for both undeformed and deformed samples to account for the different scattering mechanisms.

Calculation results agreed well with the data fitting results of measurement for an example sample of screw dislocations [4] before deformation and after 8% deformation, as shown in Fig. 6.2. Here the parameters in the relaxation times, including specularity p in phonon-boundary scattering, E in phonon-electron scattering and dislocation density  $N_d$  in phonon-dislocation scattering, are extracted from fitting the experimental data using Eq. 2.4. This data extraction process in Eq. 2.4 is to obtain the lattice thermal conductivity from the total thermal conductivity to compare with the calculation results, and it is shown in Chapter 3 of the extended model by explicitly including the phonon-dislocation scattering. Phonon peak thermal conductivity  $k_{pp}$  decreases by about 50% and the phonon peak temperature  $T_{pp}$  moves from 1.8 K before deformation to about 2 K after 8% deformation.



Figure 6.2: Comparision between the calculation results and the data fitting results for an example sample of screw dislocations [4] for undeformed state and 8% tensile deformation. The group velocity is taken as  $v_g = d\omega/dq$ , the dislocation density is calculated based on the fitting using Eq. 2.4.

This calculation was performed for another example of predominantly screw dislocations [4]. The calculation results agreed well with the data fitting results of measurement for different levels of deformation, including undeformed, 3% deformation, 7.3% deformation, and 10.3% deformation, as shown in Fig. 6.3. Here, the parameters in the relaxation times, including specularity p in phonon-boundary scattering, E in phonon-electron scattering and dislocation density  $N_d$  in phonon-dislocation scattering, are extracted from fitting the experimental data using Eq. 2.4. A phonon peak appears at about 1.8 K for the undeformed state, and it shifts to 1.9 K after 3% deformation, 2 K after 7.3% deformation, and 2.2 K after 10.3% deformation. In addition,  $k_{pp}$  is about 60 W m<sup>-1</sup> K<sup>-1</sup>before deformation, it decreases to about 40 W m<sup>-1</sup> K<sup>-1</sup>after 3% deformation, it further decreases to about 17 W m<sup>-1</sup> K<sup>-1</sup>after 7.3% deformation and about 8 W m<sup>-1</sup> K<sup>-1</sup>after 10.3%



Figure 6.3: Comparision between the calculation results and the data fitting results for another example sample of screw dislocations [4]for undeformed state and several levels of deformation. The group velocity is taken as  $v_g = d\omega/dq$ , the dislocation density is calculated based on the fitting using Eq. 2.4.

deformation.

Calculations of the lattice thermal conductivity of materials with of predominantly edge dislocations were performed for the two samples from reference [4], separately. The calculation results agreed well with the data fitting results of measurement at different levels of deformation, including undeformed and 4% deformation for sample H2 and undeformed, 1% deformation, 2.4% deformation for sample K10, as shown in Fig. 6.4 and Fig. 6.5. Here, the parameters in the relaxation times are also extracted from fitting the experimental data using Eq. 2.4 to obtain the lattice thermal conductivity. The phonon peak is at about 1.8 K for undeformed state, and it shifts to 1.9 K after 4% deformation for sample H2 or after 1% deformation for sample K10. The  $T_{pp}$  moves to about



Figure 6.4: Comparision between the calculation results and the data fitting results for an example sample of edge dislocations (sample H2) [4] for the undeformed state and 4% tensile deformation. The group velocity is taken as  $v_g = d\omega/dq$ , the dislocation density was calculated based on the fitting using Eq. 2.4.

2.4 K after 2.4% deformation for sample K10. In addition,  $k_{pp}$  is about 85 W m<sup>-1</sup> K<sup>-1</sup>for sample H2 before deformation, and it decreases to about 22 W m<sup>-1</sup> K<sup>-1</sup>after 4% deformation. For sample K10, the lattice thermal conductivity for sample K10 before deformation is around 57 W m<sup>-1</sup> K<sup>-1</sup>, it decreases to be about 33 W m<sup>-1</sup> K<sup>-1</sup>after 1% deformation, and it further decreases to about 10 W m<sup>-1</sup> K<sup>-1</sup>after 2.4% deformation.

# 6.4 Conclusion

Lattice thermal conductivity dominates the total thermal conductivity at low temperatures (T < 2 K) for superconducting Nb. An enhanced closed form model based on Callaway is proposed to include the phonon dispersion relation of Nb and also consider different scattering mechanisms,



Figure 6.5: Comparision between the calculation results and the data fitting results for another example sample of screw dislocations (sample K10) [4] for the undeformed state and 1% and 2.4% tensile deformation. The group velocity is taken as  $v_g = d\omega/dq$ , the dislocation density was calculated based on the fitting using Eq. 2.4.

including phonon-boundary scattering, phonon-electron scattering, and phonon-dislocation scattering. Calculation of the lattice thermal conductivity of superconducting Nb match well with the fitting results using Eq. 2.4 for undeformed samples and deformed samples, separately. For the comparison of deformed samples, both edge dislocations and screw dislocations are considered. A phonon peak appears at around 1.8 K for undeformed samples and shifts to warmer temperature with the increase of deformation, while the lattice thermal conductivity  $k_{pp}$  decreases with the increase of deformation. Edge dislocations seem to decrease the lattice thermal conductivity more than the screw dislocations.

#### **CHAPTER 7**

#### THERMAL CONDUCTIVITY OF SUPERCONDUCTING TANTALUM

# 7.1 Introduction

Tantalum (Ta) is a part of the refractory metals group and also a superconducting metal, similar to Nb. Because Nb and Ta are found together, there is always Ta present in SRF cavities, so this provides some intrinsic interest and motivation for understanding the thermal conductivity of Ta. Ta and Nb behave like twins in nature and they have very similar physical and chemical properties. For example, both of them are body-center cubic materials and have similar thermal conductivity at room temperature. However, the transition temperature ( $T_c$ ) from normal conductor to superconductor of Ta is 4.48 K, which is about one half of that of Nb ( $T_c = 9.25$  K) [67].

Thermal conductivity of Ta also consists of the phonon and electronic contributions. A typical figure of these contributions to the total thermal conductivity of superconducting Ta is shown in Fig. 7.1. It shows that the electron contribution to the thermal conductivity of Ta is negligible below 1 K. Therefore, phonon dominated thermal conductivity (lattice thermal conductivity) can represent the total thermal conductivity in this temperature range. A number of factors, including sample size, specularity, free electrons, impurity concentration, and dislocation density  $N_d$ , determine  $k_g$ . Plastic deformation, which occurs in many manufacturing processes, increases  $N_d$ , while heat treatments can reduce  $N_d$  [8].

# 7.2 Monte Carlo simulation

Lattice thermal conductivity of superconducting Nb before and after deformation have been simulated in the previous chapter using the Monte Carlo method. It is used here model the lattice thermal conductivity of Ta. Similarly, the phonon dispersion relationship is needed for the simulation.

To model the thermal transport of Ta at low temperatures, where phonons dominate the thermal



Figure 7.1: The thermal conductivity of Ta is the sum of the contributions of electrons and phonons [3]. Below 1 K, the electronic contribution is negligible. The contribution of phonons dominates in this temperature range because of the condensation of free electrons into Cooper pairs.

Table 7.1: Parameters for the Ta samples used in the simulation [3], where the size refers to the diameter of the cylinder. All of the three Ta samples were deformed at temperature higher or equal to 295 K, therefore they are likely to have edge dislocations.

Sample	Size	RRR	Deformation T	Dislocation type
Ta 1	3.8 mm	185	420 K	Edge dislocations
Ta 2	3.3 mm	111	355 K	Edge dislocations
Ta 3	3.9 mm	60	295 K	Edge dislocations
Nb 1	3.2 mm	350	77 K	Screw dislocations
Nb 2	3.2 mm	1200	295 K	Edge dislocations
Nb 3	3.9 mm	250	195 K	Screw dislocations

conductivity, the phonon dispersion relation of Ta is plotted in Fig. 7.2 [15]. The samples used for comparing with the simulation results are listed in Table 7.1, where the conditions before deformation and after several levels of deformation are described for samples Ta 1, Ta 2, and Ta 3.

The relaxation times used in the simulation are similar to those of Nb. That is, for undeformed


Figure 7.2: Phonon dispersion relation of Ta in the [1 0 0] direction, replotted from [15],  $\epsilon = aq/\pi$ , where q is the wave vector,  $\epsilon$  is the reduced wave vector, a is the lattice parameter, which refers to the physical dimension of a unit cell in a crystal lattice. For Ta,  $a \approx 3.3 \times 10^{-10}$  m, the same as Nb, LA refers to longitudinal acoustic phonons, and TA refers to transverse acoustic phonons.

sample, the phonon-boundary scattering and phonon-electron scattering are considered, but the phonon-dislocation scattering is neglected. For samples after deformation, all of the three scattering mechanisms, including phonon-boundary, phonon-electron, and phonon-dislocation scattering, are considered in the simulation.

Simulation results of the lattice thermal conductivity of undeformed Ta and samples after several levels of deformation are compared using measurements in Figs. 7.3-7.5, separately. Measurements were fitted using Eq. 2.19 for undeformed samples and Eq. 2.4 for samples after deformation, and the phonon contributions are shown for the lattice thermal conductivity of Ta. The phonon peak thermal conductivity for Ta typically appears at around 1 K [4], which is approximately half that of



Figure 7.3: Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 1 with the fitting results using Eq. 2.4 from [3], from undeformed to 2%, 3.1%, and 7.3% deformation.

Nb. The simulation results agree well with the fitted data for undeformed samples Ta 1, Ta 2, and Ta 3 when including phonon-boundary scattering, phonon-electron scattering, but neglecting phonondislocation scattering. For samples after deformation, *i.e.*, 2% deformation and 3.1% deformation for sample Ta 1, 0.5%, 1.2%, 2.4%, 3.9%, and 6.2% deformation for Ta 2, and 1% deformation for sample Ta 3, the simulation results also match well with the fitted data. The phonon peak occurs at around 0.9 K in the simulation for all of the three samples Ta 1, Ta 2, and Ta 3 before deformation and it shifts to about 1 K after 2% deformation for Ta 1, 0.5% deformation for Ta 2, and 1% deformation for Ta 3. After 3.1% deformation for sample Ta 1 and 1.2% deformation for Ta 2, the peaks move to around 1.1 K. The phonon peak is broad and low after 7.3% deformation for Ta 1 and after 2.4% deformation for Ta 2. The reason for the existence of a phonon peak after large deformation



Figure 7.4: Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 2 with the fitting results using Eq. 2.4 from [3], from undeformed to 0.5%, 1.2%, 2.4%, 3.9%, and 6.2% deformation.

is because of the phonon contributions only, the total thermal conductivity will lose the phonon peak after large deformation (*e.g.*, 7.3% deformation for Ta 1). Lattice thermal conductivity at temperatures lower than the phonon peak temperature is dominated by phonon-boundary scattering and follows the  $T^3$  relation. At temperatures above the phonon peak, phonon-dislocation scattering is taking over with larger amounts of deformation. Phonon-electron scattering is contributing to the lattice thermal conductivity after 1 K and will dominate the conductivity after 1.5 K. These results are similar to that of Nb for the contributions from different scattering mechanisms.



Figure 7.5: Comparison between the Monte Carlo simulation results of lattice thermal conductivity of Ta 3 with the fitting results using Eq. 2.4 from [3], from undeformed to 1% deformation.

# 7.3 Comparison of total k between Nb and Ta

It has been demonstrated that the model of Eq. 2.4 matches better with the experimental data than Eq. 2.19 of K&B model [41], it is used to model the thermal conductivity of both Nb and Ta in the following.

Comparison of data from [4, 16] with fitting of the model (Eq. 2.4) are shown in Fig. 7.6 for superconducting Nb and superconducting Ta, both before and after deformation. The figures show that after deformation, thermal conductivity of both Nb and Ta decrease because of the increase of dislocation density, where the temperatures at the phonon peak ( $T_{pp}$ ) increase with deformation for both Nb and Ta. The phonon peak disappears after a certain amount of deformation. For example, the phonon peak disappears after 7.3% deformation for these Nb and Ta samples. After 10.3%



Figure 7.6: Comparison between fitting with the experimental data of the thermal conductivity of undeformed and deformed superconducting Nb [4] and Ta [16]. The dislocation density is calculated during the fitting processes.

deformation for this Nb sample, the thermal conductivity increases proportionally with temperature. This temperature relation is not with  $T^3$  because both boundary scattering and dislocation scattering are important to contribute to the lattice thermal conductivity, and boundary scattering contributes to  $k_g$  as  $T^3$  while dislocation scattering contributes to  $k_g$  as  $T^2$ . Another interesting similarity between Nb and Ta is that  $k_{pp}$  occurs at  $T/T_c \approx 0.2$  and  $k_{lm}$  (local minimum) occurs at  $T/T_c \approx 0.3$ for both Nb and Ta. However, at  $T/T_c \approx 0.4$ , the thermal conductivity of Ta is greater than the value at the phonon peak, while the thermal conductivity of Nb is smaller than the value at its phonon peak.

## 7.3.1 Comparison of simulated lattice k between undeformed Nb and Ta

SRF cavities can be formed using large grain Nb sheet. These Nb sheets are deformed and then welded to make the cavities and then heat treated to get rid of the impurities and also the dislocations. Before deformation, the dislocation density is usually low and the deformation will increase the dislocation density.

Monte Carlo simulation results of the lattice thermal conductivity of superconducting Nb and Ta samples before deformation are shown in Fig. 7.7 by comparing with the model of Eq. 2.4. The simulation results match well with the fitted model by including the effect of phonon-



Figure 7.7: Comparison between Monte Carlo simulation of the lattice thermal conductivity with the fitting results using Eq. 2.4 for undeformed Nb [4] and undeformed Ta [16]. The parameters, including size, RRR, deformation temperature, and the calculated dislocation density, of the Nb and Ta samples are listed in Table 4.1.

boundary scattering, phonon-electron scattering, and phonon-dislocation scattering. Here the phonon-dislocation scattering is less important for undeformed samples with the other two scattering mechanisms because of the low dislocation density before deformation. The phonon peak appears at about 1.8 K for Nb and 0.9 K for Ta for all of the three samples. The temperature at the phonon peak is close to  $0.2T_c$ . Therefore the phonon peak at the undeformed Nb and Ta has small variations.

## 7.3.2 Comparison of simulated lattice *k* between deformed Nb and Ta

At undeformed states, the dislocation density is usually low so that it will not affect the lattice thermal conductivity effectively compared with the other scattering mechanisms, *i.e.*phonon-boundary scattering and phonon-dislocation scattering. However, the dislocation density will increase with deformation and becomes more and more important in its effect on the lattice thermal conductivity.

Simulation results of the lattice thermal conductivity of superconducting Nb and Ta samples after deformation are shown in Fig. 7.8 by comparing with the model of Eq. 2.4. The simulation results also match well with the fitting model by including the effect of phonon-boundary scattering, phonon-electron scattering and phonon-dislocation scattering. Here the phonon-dislocation scattering is more important for deformed samples than the other two scattering mechanisms, due



Figure 7.8: Comparison between Monte Carlo simulation of the lattice thermal conductivity with the fitting results using Eq. 2.4 of undeformed and deformed Nb [4] and Ta [16]. The parameters, including size, RRR, deformation temperature, and the calculated dislocation density, of the Nb and Ta samples are listed in Table 4.1.

to the increase of dislocation density after deformation. Wasserbäch observed that sample Nb1 has predominantly screw dislocations while Nb2 has predominantly edge dislocations. He also found low T (T<295 K) deformation leads to predominant screw dislocations and intermediate temperature (T>295 K) results in predominant edge dislocations [4]. As the Ta samples were deformed at T>295 K, which falls into the intermediate temperature range, they are likely to have edge dislocations [16]. It is shown from the figure that  $k_{pp}$  decreases with deformation and will eventually disappear after large deformation, while  $T_{pp}$  increases with deformation for both Nb and Ta. These results can account for the effect of dislocations. It can be also seen that the lattice thermal conductivity of Nb has a greater value than Ta in similar conditions. Also, for a given deformation, the ratio of the thermal conductivity to the undeformed state ( $k/k_{\epsilon=0}$ ) for Ta is smaller than that in Nb.

## 7.4 Conclusion

Monte Carlo solutions of the BTE have been successfully performed to model the lattice thermal conductivity of superconducting Ta before and after deformation given the phonon dispersion relationship of Ta. Different scattering mechanisms, including phonon-boundary, phonon-electron, and phonon-dislocation scattering, which contribute to the lattice thermal conductivity of Ta,

were used in the simulation to calculate the lattice thermal conductivity of Ta for samples before and after several deformations. The simulation results agree well with the results extracted from experiments [3] for different samples both before and after deformation. The results show that boundary scattering dominates for temperatures smaller than 0.5 K, where the phonon mean free path is comparable to the size of the sample, and that phonon-electron scattering is important when the temperature is greater than 1 K and dominates the conductivity for temperatures greater than 1.3 K. Phonon-dislocation scattering affects the lattice thermal conductivity around the phonon peak temperatures (between 0.7 K and 1.1 K) and large amount of deformation leads to a lower and broader peak in this temperature range. The phonon peak appears at temperatures of approximately 0.9 K in undeformed samples and shifts to higher temperature after small amount of deformation. Phonon-dislocation scattering decreases the lattice thermal conductivity and a large amount of deformation destroys the phonon peak.

#### **CHAPTER 8**

### CONCLUSIONS

An extended model based on that of the Koechlin and Bonin model [41] has demonstrated that dislocation density  $N_d$  needs to be considered explicitly, especially in deformed Nb (a type II superconductor) and Ta (a type I superconductor). This model is based on BCS [39] (and BRT [42]) theory, and the dislocation term is based on the work of Klemens [44]. Examining the thermal conductivity of Nb and Ta using this extended model suggests that normal electrons determine k for  $T \ge 0.3T_c$ , while from  $0.1T_c \le T \le 0.25T_c$ , phonons scattered by dislocation dominate k, and phonons scattered by boundaries dominate k for  $T \le 0.1T_c$ . The extended model also shows that there are threshold dislocation densities for Nb and Ta. Below these thresholds, the contributions from dislocation scattering of phonons can be neglected in the calculation of lattice k even in the range of  $0.1T_c \le T \le 0.25T_c$ . Furthermore, k demonstrates a local maximum, often called the phonon peak  $k_{pp}$  at a temperature  $T_{pp}$ . The magnitude of  $k_{pp}$  decreases with increasing  $N_d$  while  $T_{pp}$  increases with increasing  $N_d$ , until  $k_{pp}$  disappears.

Although k is correlated with RRR for  $T \ge 0.3T_c$  (specifically k(4.2 K) = RRR/4), a few measurements [4, 8] have suggested that  $k_{pp}$  may be correlated with RRR. See, for example, Fig. 2.5. This result is unexpected based on the BRT model of k of superconductors [42] and the theory of metals [37]. The extended model offers an explanation based on the results of Wasserbäch [4] and Chandrasekaran [8]. The coefficient of the phonon-boundary scattering term varies with RRR. In samples with large  $k_{pp}$ , the dislocations have little effect and the value of the  $k_{pp}$  is a result of the balance between phonon-boundary and phonon-electron scattering.

The Callaway model [95] for the lattice thermal conductivity has been extended to include a phonon dispersion relation. This allows consideration of variable phonon group velocities. However, this model is limited by the requirement that all of the relaxation times being considered to be of the same form. The Callaway model is also most suited to simple regular geometries, *e.g.*, rectangular, spherical, or cylindrical geometries. Hence, an energy-based, variance-reduced Monte Carlo solution [126] of the BTE for phonons has been developed, incorporating a relaxation time approximation and the phonon dispersion relation. The Monte Carlo simulation has the advantage of being able to model the scattering mechanisms individually, as well as the possibility to be applied to more complex geometries. Simulations of lattice k of undeformed and deformed Nb and Ta match well with the results of the extended Koechlin and Bonin model [41] (Eq. 2.4) when including the relaxation times for phonon-boundary, phonon-electron, and phonon-dislocation scattering. The Monte Carlo simulation has also be used to investigate the effect on the lattice k of saturating Nb with hydrogen. The simulation results match well with measured results for Nb saturated with H, bolstering the argument that H impurity affect the lattice k at near saturation.

Future work includes measuring the dislocation density of predominant screw and edge dislocations to verify using the extended K& B model (Eq. 2.4) to estimate the dislocation density for various levels of deformation. It will be also important to apple the developed Monte Carlo simulation method to superconducting thin film Nb and other superconductors with higher critical temperature, *e.g.*. Nb3Sn and NbN. After this, measuring the thermal conductivity of Nb with different levels of deformation and also Nb thin films will greatly help verify the extended model and the Monte Carlo simulation. APPENDIX

# A.1 Main program

```
1 clc; %% Clear
2 tic; %% Calculate computing time
3 %%%%%%%%%%% Parameters
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq Et; %% Global values
5 l=9.75*10^(-5); a=5*10^(-6); b=5*10^(-6); % dimension of the cell ...
     unit: m
6 V=l*a*b; % Volume, consider a rectangle
7 A=a*b; % Surface area of the first and last wall (consider isothermal)
8 h=6.63*10^(-34); % Planck constant
9 hbar=h/(2*pi); % Reduced Planck constant
10 kb=1.38*10^(-23); % Boltzmann constant
11 vs=[909 384.6]*2*pi; % sound velocity in LA and TA phonons
12 c=[-3.04*10^(-8) -5.52*10^(-10)]*2*pi; % constant to calculate velocity
13 W=0.3*10^(-17); % scaling factor
14 qmax=1.9*10^10; % Maximum Wavenumber
15 wmax=[6.795*10^12; 7.1075*10^12]*2*pi; % Maximum Frequency
16 qh=0.5*qmax; % Half maximum Frequency
17 wth=vs(2)*qh+c(2)*qh^2; % w1/2=w(1/2*qmax) for TA phonons
18 dw=wmax(2)/1000; % Frequency intervel
19 g=[1 2]; % Degenarations, LA or TA
20 Th=2.05; % Hot side Temperature
21 Tc=1.95; % Cold side Temperature
22 Teq=2.0; % Reference Temperature
23 for i=1:956
      w1(i)=i*dw; % Discreate frequency LA 1000 intervels (1-956)
24
25 end
26 for i=957:1:1000
  w1(i)=wmax(1)-(i-956)*dw; % intervels (957-1000)
27
28 end
```

```
29 for i=1:1000
30
      w2(i)=i*dw; % Discreate frequency TA 1000 intervels
31 end
32 dt=1*10^(-8); % time step
33 for i=1:40
      Td(i) =- (Th-Tc)/39*i+Th+(Th-Tc)/39; % Diffusion limit of T profile
34
      Tb(i)=((Th^4+Tc^4)/2)^0.25; % Ballistic limit of T profile
35
36 end
37 %%%%%%%%% end of parameters
38 %%%%%%%%%%%% Initialization
39 for i=1:1:956
40
      q1(i) = (-vs(1) + sqrt(vs(1)^2+4*c(1)*w1(i)))/(2*c(1)); % Wavenumber ...
          distribution 1-956
41
      v1(i)=vs(1)+2*c(1)*q1(i); % Group Velocity distribution 1-956
42 end
43 for i=957:1:1000
      q1(i)=(-vs(1)-sqrt(vs(1)^2+4*c(1)*w1(i)))/(2*c(1)); % Wavenumber ...
44
          distribution 957-1000
      v1(i)=abs(vs(1)+2*c(1)*q1(i)); % Wavenumber distribution 957-1000
45
46 end
47 for i=1:1000
48
      q2(i)=(-vs(2)+sqrt(vs(2)^2+4*c(2)*w2(i)))/(2*c(2)); % Wavenumber ...
          distribution 1-1000
      v2(i)=vs(2)+2*c(2)*q2(i); % Velocity distribution 1-1000
49
50 end
51 %%%%%%%%%%%% hot side initialization
52 for i=1:1000
      nlh(i)=hbar*wl(i)*(1/W)*V*(1/(exp((hbar*wl(i)/(kb*Th)))-1)-
53
54
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i)^2)/(2*pi^2*v1(i)))*dw;
                                                                             . . .
          % LA number for hot side, positive energy
      nth(i)=hbar*w2(i)*(1/W)*V*(1/(exp((hbar*w1(i)/(kb*Th)))-1)-
55
56
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q2(i)^2)/(2*pi^2*v2(i)))*dw*2;
                                                                            . . .
          % TA number for hot side, positive energy
```

```
57
       nh(i)=nlh(i)+nth(i); % Total number for hot side
       Ph(i)=nlh(i)/nh(i); % probability of LA for hot side
58
59 end
60 Nhh=sum(nh);
61 Nh=floor(sum(nh)); % number of phonons for hot side
62 Mh(1)=nh(1);
63 Fh (1) = 0;
64 for i=2:1:1000
      Mh(i)=Mh(i-1)+nh(i); % summation of ith phonon numbers
65
       Fh(i)=Mh(i)/Nhh; % determine which intervel
66
67 end
68 %%%%%%%%%% Hot side only has one cell
69 for j=1:Nh
           R=rand; % random number
70
           al=1; % frequeny invervel No. 1
71
           b1=1000; % frequeny invervel No. 1000
72
           while((R-Fh(a1))*(Fh(a1+1)-R)<0) % bisection method to ...</pre>
73
              determine which interval
74
              mm=round((a1+b1)/2); % get integer
               if (abs(R-Fh(a1)) < abs(R-Fh(b1)))
75
                   if (R>Fh(mm))
76
                       al=mm;
77
                   else
78
                       b1=mm;
79
80
                   end
               else
81
                   if (R<Fh(mm))</pre>
82
                       b1=mm;
83
84
                   else
                       al=mm;
85
                   end
86
87
               end
88
           end
```

9	R1=rand; %random number
0	R2=rand; %random number
1	f(j)=w1(a1)+(2*R2-1)*dw/2; % bisection method to determine
	which interval
2	if (R1 <ph(a1)) %="" (la)<="" polarization="" td=""></ph(a1))>
3	ml(j)=(-vs(1)+sqrt(vs(1)^2+4*c(1)*f(j)))/(2*c(1)); % LA
	wavevector (assume isotropy)
4	<pre>mt(j)=0; % TA wavevector=0</pre>
5	<pre>vl(j)=vs(1)+2*c(1)*ml(j); % LA magnitude group velocity</pre>
6	vt(j)=0; % TA velocity=0
7	else % Polarization (TA)
8	<pre>mt(j)=(-vs(2)+sqrt(vs(2)^2+4*c(2)*f(j)))/(2*c(2)); % TA</pre>
	wavevector (assume isotropy)
9	<pre>ml(j)=0; % LA wavevector=0</pre>
0	<pre>vt(j)=vs(2)+2*c(2)*mt(j); % TA magnitude group velocity</pre>
1	vl(j)=0; % LA velocity=0
2	end
3	v(j)=(vt(j)+vl(j)); % total velocity
4	<pre>m(j)=mt(j)+ml(j); % total wave number</pre>
5	R1=rand; % random number
6	R2=rand; % random number
7	R3=rand; % random number
8	R4=rand; % random number
9	R5=rand; % random number
0	K2=2*pi*R4; % angle 1 psi
1	K1=2*R5-1; % angle 2 cos(theta)
2	<pre>vg(j,2)=v(j)*sqrt(1-K1^2)*cos(K2); % direction of velocity</pre>
	sin(theta)*cos(psi)
3	<pre>vg(j,3)=v(j)*sqrt(1-K1^2)*sin(K2); % direction of velocity</pre>
	<pre>sin(theta)*sin(psi)</pre>
4	<pre>vg(j,1)=v(j)*K1; % direction of velocity cos(theta)</pre>
5	x1(j)=(R1)*1; % location of particles in cell 1
6	x2(j)=(R2)*a; % location of particles in cell 1

```
117
           x3(j)=(R3)*b; % location of particles in cell 1
118
           s(j)=1; % Sign of particles 1(T>Teq), -1(T<Teq)</pre>
119 end
   %%%%%%%%%%%% end of hot side initialization
120
122
   for i=1:1000
123
       nlc(i) =-hbar*w1(i) *1/W*V*(1/(exp((hbar*w1(i)/(kb*Tc)))-1)-
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i)^2)/(2*pi^2*v1(i)))*dw;
124
                                                                          . . .
          % LA number for hot side
       ntc(i) = -hbar*w2(i)*1/W*V*(1/(exp((hbar*w1(i)/(kb*Tc)))-1)-
125
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q2(i)^2)/(2*pi^2*v2(i)))*dw*2; ...
126
          % TA number for hot side
127
       nc(i)=nlc(i)+ntc(i); % Total number for hot side
       Pc(i)=nlc(i)/nc(i); % probability of LA for hot side
128
129 end
130 Ncc=sum(nc);
131 Nc=floor(sum(nc)); % number of phonons for hot side
132 Mc(1) = nc(1);
133 Fc(1) = 0;
134 for i=2:1:1000
       Mc(i)=Mc(i-1)+nc(i); % summation of ith phonon numbers
135
                         % to determine phonon belong to which intervel
136
       Fc(i)=Mc(i)/Ncc;
137 end
138 N=Nh+Nc*39; % Total number of phonons
139 %%%%%%%%%%% cold side has 39 cells
  for i=2:1:40
140
       for j=1:1:Nc
141
           R=rand; %random number
142
143
           al=1; % frequeny invervel No. 1
           b1=1000; % frequeny invervel No. 1000
144
           while((R-Fc(a1))*(Fc(a1+1)-R)<0)</pre>
145
146
               mm = round((a1+b1)/2);
```

147	<pre>if (abs(R-Fc(al))<abs(r-fc(bl))) %="" bisection="" method="" pre="" to<=""></abs(r-fc(bl)))></pre>
	determine which interval
148	if (R>Fc(mm))
149	al=mm;
150	else
151	bl=mm;
152	end
153	else
154	if (R <fc(mm))< th=""></fc(mm))<>
155	bl=mm;
156	else
157	al=mm;
158	end
159	end
160	end
161	R1=rand;
162	R2=rand;
163	f (Nh+(i-2)*Nc+j)=w1(a1)+(2*R2-1)*dw/2;
164	if (R1 <pc(al)) %="" (la)<="" polarization="" th=""></pc(al))>
165	ml(Nh+(i-2)*Nc+j)=(-vs(1)+sqrt(vs(1)^2+4*c(1)*f(Nh+(i-2)*Nc+j)))/(2*c(1));
	% LA wavevector (assume isotropy)
166	<pre>mt(Nh+(i-2)*Nc+j)=0;% TA wavevector=0</pre>
167	vl(Nh+(i-2)*Nc+j)=vs(1)+2*c(1)*ml(Nh+(i-2)*Nc+j); % LA
	magnitude group velocity
168	vt(Nh+(i-2)*Nc+j)=0; % TA velocity=0
169	else
170	mt (Nh+(i-2)*Nc+j)=(-vs(2)+sqrt(vs(2)^2+4*c(2)*f(Nh+(i-2)*Nc+j)))/(2*c(2));
	% TA wavevector (assume isotropy)
171	<pre>ml(Nh+(i-2)*Nc+j)=0; % LA wavevector=0</pre>
172	vt(Nh+(i-2)*Nc+j)=vs(2)+2*c(2)*mt(Nh+(i-2)*Nc+j); % TA
	magnitude group velocity
173	vl(Nh+(i-2)*Nc+j)=0; % LA velocity=0
174	end

1	
175	v(Nh+(i-2)*Nc+j)=vt(Nh+(i-2)*Nc+j)+vl(Nh+(i-2)*Nc+j); % total
	velocity
176	<pre>m(Nh+(i-2)*Nc+j)=mt(Nh+(i-2)*Nc+j)+ml(Nh+(i-2)*Nc+j); % total</pre>
	wave number
177	R1=rand; % random number
178	R2=rand; % random number
179	R3=rand; % random number
180	R4=rand; % random number
181	R5=rand; % random number
182	K2=2*pi*R4; % angle 1 psi
183	K1=2*R5-1; % angle 2 cos(theta)
184	vg(Nh+(i-2)*Nc+j,2)=v(Nh+(i-2)*Nc+j)*sqrt(1-K1^2)*cos(K2); %
	direction of velocity sin(theta)*cos(psi)
185	vg(Nh+(i-2)*Nc+j,3)=v(Nh+(i-2)*Nc+j)*sqrt(1-K1^2)*sin(K2); %
	direction of velocity sin(theta)*sin(psi)
186	vg(Nh+(i-2)*Nc+j,1)=v(Nh+(i-2)*Nc+j)*K1; % direction of
	velocity cos(theta)
187	<pre>x1(Nh+(i-2)*Nc+j)=(i-1+R1)*l; % location of particles in cell 2-40</pre>
188	<pre>x2(Nh+(i-2)*Nc+j)=(R2)*a; % location of particles in cell 2-40</pre>
189	x3(Nh+(i-2)*Nc+j)=(R3)*b; % location of particles in cell 2-40
190	<pre>s(Nh+(i-2)*Nc+j)=-1; % Sign of particles 1(T&gt;Teq), -1(T<teq)< pre=""></teq)<></pre>
191	end
192	end
193	%%%%%%%%% end of cold side initialization
194	%%%%%% Drift process
195	u1=0; % iteration times
196	d=0.996; % specularity
197	Ttol=1500*dt; % Total time
198	<pre>for t=dt:dt:Ttol</pre>
199	for i=1:40
200	<pre>Et(i)=0; % Total energy=0</pre>
201	NNN(i)=0; % Number of particles in each cell before drifting
202	end

203	Nd1=0; % number of particles to be deleted in cell 1 $$
204	Nd40=0; % number of particles to be deleted in cell 40
205	<pre>for j=1:1:N % drift process</pre>
206	<pre>x1(j)=x1(j)+vg(j,1)*dt; % update coordinate 1</pre>
207	<pre>x2(j)=x2(j)+vg(j,2)*dt; % update coordinate 2</pre>
208	<pre>x3(j)=x3(j)+vg(j,3)*dt; % update coordinate 3</pre>
209	if $(x2(j) \leq 0)$ % reach bottom boundary
210	R=rand; % random number
211	if (R <d) %="" reflection<="" specular="" td=""></d)>
212	x2(j) = -x2(j); % update coordinate 2
213	<pre>vg(j,2)=-vg(j,2); % particles bounce back</pre>
214	else % diffusive reflection
215	R1=rand; % random number
216	R2=rand; % random number
217	K1=2*R1-1; % angle 1
218	K2=2*pi*R2; % angle 2
219	<pre>ddt=abs((x2(j))/(vg(j,2))); % time for reflection</pre>
220	<pre>x1(j)=x1(j)-ddt*vg(j,1); % update coordinate 1</pre>
221	<pre>x3(j)=x3(j)-ddt*vg(j,3); % update coordinate 2</pre>
222	<pre>x2(j)=x2(j)-ddt*vg(j,2); % update coordinate 3</pre>
223	<pre>vg(j,1)=v(j)*K1;% velocity random direction</pre>
224	<pre>vg(j,2)=abs(v(j)*sqrt(1-K1^2)*sin(K2)); % velocity random direction</pre>
225	<pre>vq(j,3)=v(j)*sqrt(1-K1^2)*cos(K2); % velocity random</pre>
	direction
226	<pre>x1(j)=x1(j) + ddt*vg(j,1); % update coordinate 1</pre>
227	x3(j)=x3(j) + ddt*vg(j,3); % update coordinate 1
228	<pre>x2(j)=x2(j) + ddt*vg(j,2); % update coordinate 1</pre>
229	end
230	
231	<pre>elseif (x2(j)≥a) % reach top boundary</pre>
232	R=rand; % random number
233	if (R <d) %="" reflection<="" specular="" td=""></d)>
1	

1	
234	<pre>x2(j)=2*a-x2(j); % update coordinate 2</pre>
235	<pre>vg(j,2)=-vg(j,2); % particles bounce back</pre>
236	else % diffusive reflection
237	R1=rand; % random number
238	R2=rand; % random number
239	K1=2*R1-1; % angle 1
240	K2=2*pi*R2; % angle 2
241	<pre>ddt=abs((a-x2(j))/(vg(j,2))); % time for reflection</pre>
242	<pre>x1(j)=x1(j)-ddt*vg(j,1); % update coordinate 1</pre>
243	<pre>x3(j)=x3(j)-ddt*vg(j,3); % update coordinate 2</pre>
244	<pre>x2(j)=x2(j)-ddt*vg(j,2); % update coordinate 3</pre>
245	<pre>vg(j,1)=v(j)*K1; % velocity random direction</pre>
246	vg(j,2)=-abs(v(j)*sqrt(1-K1^2)*sin(K2)); % velocity
	random direction
247	<pre>vg(j,3)=v(j)*sqrt(1-K1^2)*cos(K2); % velocity random</pre>
	direction
248	<pre>x1(j)=x1(j) + ddt*vg(j,1); % update coordinate 1</pre>
249	x3(j)=x3(j) + ddt*vg(j,3); % update coordinate 1
250	<pre>x2(j)=x2(j) + ddt*vg(j,2); % update coordinate 1</pre>
251	end
252	end
253	
254	if (x3(j)≤0) % reach left boundary
255	R=rand; % random number
256	<pre>if (R<d) %="" pre="" reflection<="" specular=""></d)></pre>
257	<pre>x3(j)=-x3(j); % update coordinate 3</pre>
258	<pre>vg(j,3)=-vg(j,3); % particles bounce back</pre>
259	else % diffusive reflection
260	R1=rand; % random number
261	R2=rand; % random number
262	K1=2*R1-1; % angle 1
263	K2=2*pi*R2; % angle 2
264	<pre>ddt=abs(x3(j)/(vg(j,3))); % time for reflection</pre>

265	<pre>x1(j)=x1(j)-ddt*vg(j,1); % update coordinate 1</pre>
266	<pre>x3(j)=x3(j)-ddt*vg(j,3); % update coordinate 2</pre>
267	<pre>x2(j)=x2(j)-ddt*vg(j,2); % update coordinate 3</pre>
268	<pre>vg(j,1)=v(j)*K1; % velocity random direction</pre>
269	<pre>vg(j,2)=v(j)*sqrt(1-K1^2)*sin(K2); % velocity random</pre>
	direction
270	vg(j,3)=abs(v(j)*sqrt(1-K1^2)*cos(K2)); % velocity
	random direction
271	<pre>x1(j)=x1(j) + ddt*vg(j,1); % update coordinate 1</pre>
272	<pre>x3(j)=x3(j) + ddt*vg(j,3); % update coordinate 1</pre>
273	<pre>x2(j)=x2(j) + ddt*vg(j,2); % update coordinate 1</pre>
274	end
275	
276	elseif (x3(j)≥b) % reach right boundary
277	R=rand; % random number
278	<pre>if (R<d) %="" pre="" reflection<="" specular=""></d)></pre>
279	<pre>x3(j)=2*b-x3(j); % update coordinate 3</pre>
280	<pre>vg(j,3)=-vg(j,3); % particles bounce back</pre>
281	else % diffusive reflection
282	R1=rand; % random number
283	R2=rand; % random number
284	K1=2*R1-1; % angle 1
285	K2=2*pi*R2; % angle 2
286	<pre>ddt=abs((b-x3(j))/(vg(j,3))); % time for reflection</pre>
287	<pre>x1(j)=x1(j)-ddt*vg(j,1); % update coordinate 1</pre>
288	<pre>x3(j)=x3(j)-ddt*vg(j,3); % update coordinate 2</pre>
289	<pre>x2(j)=x2(j)-ddt*vg(j,2); % update coordinate 3</pre>
290	<pre>vg(j,1)=v(j)*K1; % velocity random direction</pre>
291	vg(j,2)=v(j)*sqrt(1-K1^2)*sin(K2); % velocity random
	direction
292	vg(j,3)=-abs(v(j)*sqrt(1-K1^2)*cos(K2)); % velocity
	random direction
293	<pre>x1(j)=x1(j) + ddt*vg(j,1); % update coordinate 1</pre>

```
294
                     x3(j)=x3(j) + ddt*vg(j,3); % update coordinate 1
                     x2(j)=x2(j) + ddt*vg(j,2); % update coordinate 1
295
                end
296
297
            end
298
            if (x1(j)<l) % reach left (delete this phonon)</pre>
299
300
                Nd1=Nd1+1; % put the deleted particle in cell 1
            elseif (x1(j)>39*1) % reach right (delete this phonon)
301
302
                Nd40=Nd40+1; % put the deleted particle in cell 40
303
            end
304
            Ndd=Nd1+Nd40; % total particle to be deleted
305
        end
306 %%%%%%%%%%%%%%%%%% end of drift process
307 %%%%% reinitialization
   %%%%%%%%%%%%%%%%%% Determine the properties of hot cell
308
        for j=1:Nh
309
            R=rand; % random number
310
            a1=1; % frequeny invervel No. 1
311
            b1=1000; % frequeny invervel No. 1000
312
            while ((R-Fh(a1)) * (Fh(a1+1)-R) < 0)
313
                mm=round((a1+b1)/2); % get integer
314
                if (abs(R-Fh(a1)) < abs(R-Fh(b1))) % bisection method to ...
315
                    determine which interval
                     if (R>Fh(mm))
316
317
                         al=mm;
                     else
318
                         b1=mm;
319
320
                     end
321
                else
                     if (R<Fh(mm))
322
323
                         b1=mm;
324
                     else
325
                         al=mm;
```

326	end
327	end
328	end
329	R1=rand; %random number
330	R2=rand; %random number
331	f(N+j)=w1(a1)+(2*R2-1)*dw/2; % assign the frequency
332	if (R1 <ph(a1)) %="" (la)<="" polarization="" td=""></ph(a1))>
333	<pre>ml(N+j)=(-vs(1)+sqrt(vs(1)^2+4*c(1)*f(N+j)))/(2*c(1)); % LA</pre>
	wavevector (assume isotropy)
334	<pre>mt(N+j)=0; % TA wavevector=0</pre>
335	<pre>vl(N+j)=vs(1)+2*c(1)*ml(N+j); % LA magnitude group velocity</pre>
336	vt(N+j)=0; % TA velocity=0
337	else % Polarization (TA)
338	<pre>mt(N+j)=(-vs(2)+sqrt(vs(2)^2+4*c(2)*f(N+j)))/(2*c(2)); % TA</pre>
	wavevector (assume isotropy)
339	<pre>ml(N+j)=0; % LA wavevector=0</pre>
340	<pre>vt(N+j)=vs(2)+2*c(2)*mt(N+j); % TA magnitude group velocity</pre>
341	vl(N+j)=0; % LA velocity=0
342	end
343	v(N+j)=vt(N+j)+vl(N+j); % total velocity
344	<pre>m(N+j)=mt(N+j)+ml(N+j); % total wave number</pre>
345	R1=rand; % random number
346	R2=rand; % random number
347	R3=rand; % random number
348	R4=rand; % random number
349	R5=rand; % random number
350	K2=2*pi*R4; % angle 1 psi
351	K1=2*R5-1; % angle 2 cos(theta)
352	vg(N+j,2)=v(N+j)*sqrt(1-K1^2)*cos(K2); % direction of velocity
	<pre>sin(theta)*cos(psi)</pre>
353	vg(N+j,3)=v(N+j)*sqrt(1-K1^2)*sin(K2); % direction of velocity
	<pre>sin(theta)*sin(psi)</pre>
354	<pre>vg(N+j,1)=v(N+j)*K1; % direction of velocity cos(theta)</pre>

355 x1(N+j)=R1\*1; % location of particles in cell 1 x2(N+j)=R2\*a; % location of particles in cell 1 356 x3(N+j)=R3\*b; % location of particles in cell 1 357 358 s(N+j)=1; % Sign of particles 1(T>Teq), -1(T<Teq)</pre> 359 end <u> ୧</u>୧୧୧୧୧୧ Determine the properties of cold wall 360 361 for j=1:Nc R=rand; % random number 362 a1=1; % frequeny invervel No. 1 363 b1=1000; % frequeny invervel No. 1000 364 while((R-Fc(a1))\*(Fc(a1+1)-R)<0) % bisection method to ...</pre> 365 determine which interval mm=round((a1+b1)/2); % get integer 366 367 if (abs(R-Fc(al)) < abs(R-Fc(bl))) % bisection method if (R>Fc(mm)) 368 369 al=mm; 370 else 371 b1=mm; 372 end 373 else 374 if (R<Fc(mm)) 375 b1=mm; else 376 377 al=mm; 378 end 379 end end 380 R1=rand; %random number 381 382 R2=rand; %random number f(N+Nh+j)=w1(a1)+(2\*R2-1)\*dw/2; % assign the frequency 383 if (R1<Pc(a1)) % Polarization (LA)</pre> 384 385  $ml(N+Nh+j) = (-vs(1)+sqrt(vs(1)^2+4*c(1)*f(N+Nh+j)))/(2*c(1)); \dots$ % LA wavevector (assume isotropy)

<pre>mt(N+Nh+j)=0; % TA wavevector=0</pre>
<pre>vl(N+Nh+j)=vs(1)+2*c(1)*ml(N+Nh+j); % LA magnitude group</pre>
velocity
<pre>vt(N+Nh+j)=0; % TA velocity=0</pre>
else % Polarization (TA)
<pre>mt(N+Nh+j)=(-vs(2)+sqrt(vs(2)^2+4*c(2)*f(N+Nh+j)))/(2*c(2));</pre>
% TA wavevector (assume isotropy)
<pre>ml(N+Nh+j)=0; % LA wavevector=0</pre>
<pre>vt(N+Nh+j)=vs(2)+2*c(2)*mt(N+Nh+j); % TA magnitude group</pre>
velocity
vl(N+Nh+j)=0; % LA velocity=0
end
v(N+Nh+j)=(vt(N+Nh+j)+vl(N+Nh+j)); % total velocity
<pre>m(N+Nh+j)=mt(N+Nh+j)+ml(N+Nh+j); % total wave number</pre>
R1=rand; % random number
R2=rand; % random number
R3=rand; % random number
R4=rand; % random number
R5=rand; % random number
K2=2*pi*R4; % angle 1 psi
K1=2*R5-1; % angle 2 cos(theta)
vg(N+Nh+j,2)=v(N+Nh+j)*sqrt(1-K1^2)*cos(K2); % direction of
<pre>velocity sin(theta)*cos(psi)</pre>
vg(N+Nh+j,3)=v(N+Nh+j)*sqrt(1-K1^2)*sin(K2); % direction of
<pre>velocity sin(theta)*sin(psi)</pre>
<pre>vg(N+Nh+j,1)=v(N+Nh+j)*K1; % direction of velocity cos(theta)</pre>
<pre>x1(N+Nh+j)=(39+R1)*l; % location of particles in cell 40</pre>
<pre>x2(N+Nh+j)=(R2)*a; % location of particles in cell 40</pre>
<pre>x3(N+Nh+j)=(R3)*b; % location of particles in cell 40</pre>
<pre>s(N+Nh+j)=-1; % Sign of particles 1(T&gt;Teq), -1(T<teq)< pre=""></teq)<></pre>
end
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
Nmax=N+Nh+Nc; % maximum number of particles

```
N=Nmax-Ndd; % real number of particles
414
        i=0; % indication of which particle to be reassigned
415
        if (Ndd \leq (Nh+Nc)) % If deleted particles \leq reassigned particles
416
417
            for j=1:N
                if (x1(j) < 1 | | x1(j) > 39 \times 1) % if phonons are out of boundary ...
418
                    cell, reassign the reinitialized phonons to them
419
                     x1(j)=x1(Nmax-i); % update location
420
                     x2(j)=x2(Nmax-i); % update location
                     x3(j)=x3(Nmax-i); % update location
421
422
                     vq(j,1)=vq(Nmax-i,1); % update velocity 1
423
                     vg(j,2)=vg(Nmax-i,2); % update velocity 2
                     vg(j,3)=vg(Nmax-i,3); % update velocity 3
424
                     f(j)=f(Nmax-i); % update frequency
425
426
                     ml(j)=ml(Nmax-i); % update wave numeber LA
                     mt(j)=mt(Nmax-i); % update wave numeber TA
427
                     vl(j)=vl(Nmax-i); % update velocity LA
428
                     vt(j)=vt(Nmax-i); % update velocity TA
429
                     s(j)=s(Nmax-i); % update sign of particles
430
                     m(j)=ml(j)+mt(j); % update total wave number (either LA ...
431
                        or TA)
                     v(j)=vl(j)+vt(j); % update total velocity (either LA or TA)
432
433
                     i=i+1;
                end
434
435
            end
        else % If deleted particles > reassigned particles
436
437
            k=1;
            for j=1:N
438
                if (x1(j) < 1 | | x1(j) > 39 \times 1) % if phonons are out of boundary ...
439
                    cell, reassign the reinitialized phonons to them
                     if (i \le (Nh+Nc)) % If the reassigned particles \le Nh+Nc
440
                         x1(j)=x1(Nmax-i); % update location
441
                         x2(j)=x2(Nmax-i); % update location
442
443
                         x3(j)=x3(Nmax-i); % update location
```

444	<pre>vg(j,1)=vg(Nmax-i,1); % update velocity 1</pre>
445	<pre>vg(j,2)=vg(Nmax-i,2); % update velocity 2</pre>
446	<pre>vg(j,3)=vg(Nmax-i,3); % update velocity 3</pre>
447	<pre>f(j)=f(Nmax-i); % update frequency</pre>
448	<pre>ml(j)=ml(Nmax-i); % update wave numeber LA</pre>
449	<pre>mt(j)=mt(Nmax-i); % update wave numeber TA</pre>
450	vl(j)=vl(Nmax-i); % update velocity LA
451	<pre>vt(j)=vt(Nmax-i); % update velocity TA</pre>
452	<pre>s(j)=s(Nmax-i); % update sign of particles</pre>
453	<pre>m(j)=ml(j)+mt(j); % update total wave number</pre>
	(either LA or TA)
454	<pre>v(j)=vl(j)+vt(j); % update total velocity (either</pre>
	LA or TA)
455	i=i+1;
456	<pre>else % If the reassigned particles &gt; Nh+Nc</pre>
457	while(x1(N+k) <l <math="">   x1(N+k)&gt;39*1) % if the rest</l>
	phonons are out of boundary cell, reassign the
	reinitialized phonons to them
458	k=k+1;
459	end
460	<pre>x1(j)=x1(N+k); % update location</pre>
461	x2(j)=x2(N+k); % update location
462	x3(j)=x3(N+k); % update location
463	<pre>vg(j,1)=vg(N+k,1); % update velocity 1</pre>
464	<pre>vg(j,2)=vg(N+k,2); % update velocity 2</pre>
465	<pre>vg(j,3)=vg(N+k,3); % update velocity 3</pre>
466	<pre>f(j)=f(N+k); % update frequency</pre>
467	<pre>ml(j)=ml(N+k); % update wave numeber LA</pre>
468	<pre>mt(j)=mt(N+k); % update wave numeber TA</pre>
469	<pre>vl(j)=vl(N+k); % update velocity LA</pre>
470	<pre>vt(j)=vt(N+k); % update velocity TA</pre>
471	<pre>s(j)=s(N+k); % update sign of particles</pre>

```
472
                       m(j)=ml(j)+mt(j); % update total wave number ...
                           (either LA or TA)
                       v(j)=vl(j)+vt(j); % update total velocity (either ...
473
                          LA or TA)
                       k=k+1;
474
475
                       i=i+1;
476
                   end
477
               end
478
           end
479
       end
   %%%%%%%%% Calculate number of energy in each cell
480
       for j=1:N
481
           for i=1:40
482
               if (x1(j) > (i-1) * l \&\& x1(j) < (i) * l)
483
                   NNN(i)=NNN(i)+s(j); % number of particles (energy) in ...
484
                      each cell
485
               end
486
           end
487
       end
   %%%%%%%%% Calculate target energy for each cell
488
       for i=1:40
489
           Et(i)=NNN(i)*W; % target energy of each cell
490
491
       end
   %%%%%%% end of reassignment of the phonons
492
   493
       T1(1)=100; % initial temperature (T0) of cell 1 iteration 1
494
       T1(2)=T1(1)-z1(T1(1))/(z1dot(T1(1))); % T0 of cell 1 iteration 2
495
       j=1;
496
497
       while (abs(T1(j+1)-T1(j))>0.01) % Iteration process
           j=j+1;
498
           T1(j+1)=T1(j)-z1(T1(j))/(z1dot(T1(j)));
499
500
       end
501
       T(1)=T1(j+1); % converge, solve T
```

```
502
   %%%%%%% Process is similar to get the following temperatures
503
        T2(1) = 100;
504
505
        T2(2) = T2(1) - z2(T2(1)) / (z2dot(T2(1)));
        j=1;
506
        while (abs(T2(j+1)-T2(j))>0.01)
507
508
             j=j+1;
             T2(j+1) = T2(j) - z2(T2(j)) / (z2dot(T2(j)));
509
510
        end
        T(2) = T2(j+1);
511
512
        T3(1) = 100;
513
        T3(2) = T3(1) - z3(T3(1)) / (z3dot(T3(1)));
514
515
        j=1;
        while (abs(T3(j+1)-T3(j))>0.01)
516
             j=j+1;
517
             T3(j+1) = T3(j) - z3(T3(j)) / (z3dot(T3(j)));
518
519
        end
        T(3)=T3(j+1);
520
521
        T4(1) = 100;
522
        T4(2) = T4(1) - z4(T4(1)) / (z4dot(T4(1)));
523
        j=1;
524
        while (abs(T4(j+1)-T4(j))>0.01)
525
526
             j=j+1;
             T4(j+1) = T4(j) - z4(T4(j)) / (z4dot(T4(j)));
527
528
        end
        T(4) = T4(j+1);
529
530
        T5(1) = 100;
531
        T5(2) = T5(1) - z5(T5(1)) / (z5dot(T5(1)));
532
533
        j=1;
534
        while (abs(T5(j+1)-T5(j))>0.01)
```

```
535
             j=j+1;
             T5(j+1) = T5(j) - z5(T5(j)) / (z5dot(T5(j)));
536
        end
537
538
        T(5)=T5(j+1);
539
        T6(1)=100;
540
541
        T6(2) = T6(1) - z6(T6(1)) / (z6dot(T6(1)));
542
        j=1;
        while (abs(T6(j+1)-T6(j))>0.01)
543
544
             j=j+1;
545
             T6(j+1) = T6(j) - z6(T6(j)) / (z6dot(T6(j)));
546
        end
        T(6)=T6(j+1);
547
548
        T7(1) = 100;
549
        T7(2) = T7(1) - z7(T7(1)) / (z7dot(T7(1)));
550
551
        j=1;
        while (abs(T7(j+1)-T7(j))>0.01)
552
             j=j+1;
553
             T7(j+1) = T7(j) - z7(T7(j)) / (z7dot(T7(j)));
554
        end
555
        T(7) = T7(j+1);
556
557
        T8(1)=100;
558
        T8(2) = T8(1) - z8(T8(1)) / (z8dot(T8(1)));
559
        j=1;
560
        while (abs(T8(j+1)-T8(j))>0.01)
561
             j=j+1;
562
563
             T8(j+1)=T8(j)-z8(T8(j))/(z8dot(T8(j)));
564
        end
        T(8)=T8(j+1);
565
566
        T9(1) = 100;
567
```

```
568
        T9(2) = T9(1) - z9(T9(1)) / (z9dot(T9(1)));
569
        j=1;
        while (abs(T9(j+1)-T9(j))>0.01)
570
571
             j=j+1;
             T9(j+1) = T9(j) - z9(T9(j)) / (z9dot(T9(j)));
572
573
        end
574
        T(9) = T9(j+1);
575
576
        T10(1) = 100;
        T10(2) = T10(1) - z10(T10(1)) / (z10dot(T10(1)));
577
578
        j=1;
        while (abs(T10(j+1)-T10(j))>0.01)
579
             j=j+1;
580
             T10(j+1) = T10(j) - z10(T10(j)) / (z10dot(T10(j)));
581
582
        end
        T(10)=T10(j+1);
583
584
585
        T11(1) = 100;
        T11(2)=T11(1)-z11(T11(1))/(z11dot(T11(1)));
586
        j=1;
587
        while (abs(T11(j+1)-T11(j))>0.01)
588
589
             j=j+1;
             T11(j+1)=T11(j)-z11(T11(j))/(z11dot(T11(j)));
590
591
        end
        T(11) = T11(j+1);
592
593
        T12(1) = 100;
594
        T12(2) = T12(1) - z12(T12(1)) / (z12dot(T12(1)));
595
596
        j=1;
        while (abs(T12(j+1)-T12(j))>0.01)
597
             j=j+1;
598
599
             T12(j+1) = T12(j) - z12(T12(j)) / (z12dot(T12(j)));
600
        end
```

```
601
        T(12)=T12(j+1);
602
        T13(1) = 100;
603
604
        T13(2) = T13(1) - z13(T13(1)) / (z13dot(T13(1)));
        j=1;
605
        while (abs(T13(j+1)-T13(j))>0.01)
606
607
             j=j+1;
             T13(j+1) = T13(j) - z13(T13(j)) / (z13dot(T13(j)));
608
609
        end
        T(13)=T13(j+1);
610
611
        T14(1) = 100;
612
        T14(2)=T14(1)-z14(T14(1))/(z14dot(T14(1)));
613
614
        j=1;
        while (abs(T14(j+1)-T14(j))>0.01)
615
616
             j=j+1;
             T14(j+1)=T14(j)-z14(T14(j))/(z14dot(T14(j)));
617
618
        end
        T(14)=T14(j+1);
619
620
        T15(1) = 100;
621
        T15(2) = T15(1) - z15(T15(1)) / (z15dot(T15(1)));
622
        j=1;
623
        while (abs(T15(j+1)-T15(j))>0.01)
624
625
             j=j+1;
             T15(j+1) = T15(j) - z15(T15(j)) / (z15dot(T15(j)));
626
627
        end
        T(15) = T15(j+1);
628
629
        T16(1) = 100;
630
        T16(2)=T16(1)-z16(T16(1))/(z16dot(T16(1)));
631
632
        j=1;
633
        while (abs(T16(j+1)-T16(j))>0.01)
```

```
634
             j=j+1;
             T16(j+1) = T16(j) - z16(T16(j)) / (z16dot(T16(j)));
635
636
        end
637
        T(16) = T16(j+1);
638
        T17(1) = 100;
639
        T17(2) = T17(1) - z17(T17(1)) / (z17dot(T17(1)));
640
641
        j=1;
        while (abs(T17(j+1)-T17(j))>0.01)
642
643
             j=j+1;
             T17(j+1) = T17(j) - z17(T17(j)) / (z17dot(T17(j)));
644
645
        end
        T(17)=T17(j+1);
646
647
        T18(1) = 100;
648
        T18(2)=T18(1)-z18(T18(1))/(z18dot(T18(1)));
649
        j=1;
650
        while (abs(T18(j+1)-T18(j))>0.01)
651
             j=j+1;
652
             T18(j+1) = T18(j) - z18(T18(j)) / (z18dot(T18(j)));
653
        end
654
        T(18) = T18(j+1);
655
656
        T19(1) = 100;
657
        T19(2) = T19(1) - z19(T19(1)) / (z19dot(T19(1)));
658
        j=1;
659
        while (abs(T19(j+1)-T19(j))>0.01)
660
661
             j=j+1;
662
             T19(j+1) = T19(j) - z19(T19(j)) / (z19dot(T19(j)));
663
        end
        T(19)=T19(j+1);
664
665
666
        T20(1) = 100;
```

```
667
        T20(2) = T20(1) - z20(T20(1)) / (z20dot(T20(1)));
668
        j=1;
        while (abs(T20(j+1)-T20(j))>0.01)
669
670
             j=j+1;
             T20(j+1) = T20(j) - z20(T20(j)) / (z20dot(T20(j)));
671
672
        end
673
        T(20)=T20(j+1);
674
675
        T21(1) = 100;
        T21(2) = T21(1) - z21(T21(1)) / (z21dot(T21(1)));
676
677
        j=1;
        while (abs(T21(j+1)-T21(j))>0.01)
678
             j=j+1;
679
             T21(j+1) = T21(j) - z21(T21(j)) / (z21dot(T21(j)));
680
681
        end
        T(21)=T21(j+1);
682
683
684
        T22(1) = 100;
        T22(2) = T22(1) - z22(T22(1)) / (z22dot(T22(1)));
685
        j=1;
686
        while (abs(T22(j+1)-T22(j))>0.01)
687
688
             j=j+1;
             T22(j+1) = T22(j) - z22(T22(j)) / (z22dot(T22(j)));
689
690
        end
        T(22) = T22(j+1);
691
692
        T23(1) = 100;
693
        T23(2) = T23(1) - z23(T23(1)) / (z23dot(T23(1)));
694
695
        j=1;
        while (abs(T23(j+1)-T23(j))>0.01)
696
             j=j+1;
697
698
             T23(j+1) = T23(j) - z23(T23(j)) / (z23dot(T23(j)));
699
        end
```

```
700
        T(23)=T23(j+1);
701
        T24(1) = 100;
702
703
        T24(2) = T24(1) - z24(T24(1)) / (z24dot(T24(1)));
704
        j=1;
        while (abs(T24(j+1)-T24(j))>0.01)
705
706
             j=j+1;
             T24(j+1) = T24(j) - z24(T24(j)) / (z24dot(T24(j)));
707
708
        end
        T(24)=T24(j+1);
709
710
        T25(1) = 100;
711
        T25(2) = T25(1) - z25(T25(1)) / (z25dot(T25(1)));
712
713
        j=1;
        while (abs(T25(j+1)-T25(j))>0.01)
714
715
             j=j+1;
716
             T25(j+1) = T25(j) - z25(T25(j)) / (z25dot(T25(j)));
717
        end
        T(25)=T25(j+1);
718
719
720
        T26(1) = 100;
        T26(2) = T26(1) - z26(T26(1)) / (z26dot(T26(1)));
721
        j=1;
722
        while (abs(T26(j+1)-T26(j))>0.01)
723
724
             j=j+1;
             T26(j+1) = T26(j) - z26(T26(j)) / (z26dot(T26(j)));
725
726
        end
        T(26) = T26(j+1);
727
728
        T27(1) = 100;
729
        T27(2) = T27(1) - z27(T27(1)) / (z27dot(T27(1)));
730
731
        j=1;
732
        while (abs(T27(j+1)-T27(j))>0.01)
```

```
733
             j=j+1;
             T27(j+1) = T27(j) - z27(T27(j)) / (z27dot(T27(j)));
734
        end
735
736
        T(27) = T27(j+1);
737
        T28(1) = 100;
738
739
        T28(2) = T28(1) - z28(T28(1)) / (z28dot(T28(1)));
740
        j=1;
        while (abs(T28(j+1)-T28(j))>0.01)
741
742
             j=j+1;
             T28(j+1) = T28(j) - z28(T28(j)) / (z28dot(T28(j)));
743
744
        end
        T(28)=T28(j+1);
745
746
        T29(1) = 100;
747
        T29(2) = T29(1) - z29(T29(1)) / (z29dot(T29(1)));
748
749
        j=1;
        while (abs(T29(j+1)-T29(j))>0.01)
750
             j=j+1;
751
             T29(j+1) = T29(j) - z29(T29(j)) / (z29dot(T29(j)));
752
753
        end
        T(29) = T29(j+1);
754
755
        T30(1) = 100;
756
        T30(2) = T30(1) - z30(T30(1)) / (z30dot(T30(1)));
757
        j=1;
758
        while (abs(T30(j+1)-T30(j))>0.01)
759
760
             j=j+1;
761
             T30(j+1) = T30(j) - z30(T30(j)) / (z30dot(T30(j)));
762
        end
        T(30)=T30(j+1);
763
764
765
        T31(1) = 100;
```
```
766
        T31(2)=T31(1)-z31(T31(1))/(z31dot(T31(1)));
767
        j=1;
        while (abs(T31(j+1)-T31(j))>0.01)
768
769
             j=j+1;
             T31(j+1)=T31(j)-z31(T31(j))/(z31dot(T31(j)));
770
771
        end
772
        T(31)=T31(j+1);
773
774
        T32(1) = 100;
        T32(2) = T32(1) - z32(T32(1)) / (z32dot(T32(1)));
775
776
        j=1;
        while (abs(T32(j+1)-T32(j))>0.01)
777
             j=j+1;
778
             T32(j+1) = T32(j) - z32(T32(j)) / (z32dot(T32(j)));
779
        end
780
        T(32)=T32(j+1);
781
782
783
        T33(1) = 100;
        T33(2) = T33(1) - z33(T33(1)) / (z33dot(T33(1)));
784
        j=1;
785
        while (abs(T33(j+1)-T33(j))>0.01)
786
787
             j=j+1;
             T33(j+1)=T33(j)-z33(T33(j))/(z33dot(T33(j)));
788
789
        end
        T(33)=T33(j+1);
790
791
        T34(1) = 100;
792
        T34(2) = T34(1) - z34(T34(1)) / (z34dot(T34(1)));
793
794
        j=1;
        while (abs(T34(j+1)-T34(j))>0.01)
795
             j=j+1;
796
797
             T34(j+1) = T34(j) - z34(T34(j)) / (z34dot(T34(j)));
798
        end
```

```
799
        T(34)=T34(j+1);
800
        T35(1) = 100;
801
802
        T35(2) = T35(1) - z35(T35(1)) / (z35dot(T35(1)));
803
        j=1;
        while (abs(T35(j+1)-T35(j))>0.01)
804
805
             j=j+1;
             T35(j+1) = T35(j) - z35(T35(j)) / (z35dot(T35(j)));
806
807
        end
        T(35)=T35(j+1);
808
809
        T36(1) = 100;
810
        T36(2) = T36(1) - z36(T36(1)) / (z36dot(T36(1)));
811
812
        j=1;
        while (abs(T36(j+1)-T36(j))>0.01)
813
814
             j=j+1;
             T36(j+1) = T36(j) - z36(T36(j)) / (z36dot(T36(j)));
815
816
        end
        T(36)=T36(j+1);
817
818
819
        T37(1) = 100;
        T37(2) = T37(1) - z37(T37(1)) / (z37dot(T37(1)));
820
        j=1;
821
        while (abs(T37(j+1)-T37(j))>0.01)
822
823
             j=j+1;
             T37(j+1) = T37(j) - z37(T37(j)) / (z37dot(T37(j)));
824
825
        end
        T(37) = T37(j+1);
826
827
        T38(1) = 100;
828
        T38(2)=T38(1)-z38(T38(1))/(z38dot(T38(1)));
829
830
        j=1;
831
        while (abs(T38(j+1)-T38(j))>0.01)
```

```
832
            j=j+1;
833
            T38(j+1) = T38(j) - z38(T38(j)) / (z38dot(T38(j)));
834
       end
835
       T(38)=T38(j+1);
836
837
       T39(1) = 100;
       T39(2) = T39(1) - z39(T39(1)) / (z39dot(T39(1)));
838
839
       j=1;
       while (abs(T39(j+1)-T39(j))>0.01)
840
841
            j=j+1;
            T39(j+1) = T39(j) - z39(T39(j)) / (z39dot(T39(j)));
842
843
       end
       T(39)=T39(j+1);
844
845
       T40(1) = 100;
846
       T40(2) = T40(1) - z40(T40(1)) / (z40dot(T40(1)));
847
848
       j=1;
849
       while (abs(T40(j+1) - T40(j)) > 0.01)
            j=j+1;
850
            T40(j+1) = T40(j) - z40(T40(j)) / (z40dot(T40(j)));
851
852
       end
       T(40) = T40(j+1);
853
   854
   %%%%%%% Number density function for updated T
855
856
       for i=1:1:40
            for j=1:1:1000
857
                if (T(i)≥Teq)
858
                    nl(i,j)=hbar*w1(j)*1/W*V*(1/(exp((hbar*w1(j)/(kb*T(i))))-1)-
859
860
                    1/(exp((hbar*w1(j)/(kb*Teq)))-1))*((q1(j)^2)/(2*pi^2*v1(j)))*dw; ...
                        % LA number for hot side
                    nt(i,j)=hbar*w2(j)*1/W*V*(1/(exp((hbar*w2(j)/(kb*T(i))))-1)-
861
862
                    1/(exp((hbar*w2(j)/(kb*Teq)))-1))*((q2(j)^2)/(2*pi^2*v2(j)))*dw*2; ...
                        % TA number for hot side
```

```
else
863
                      nl(i, j) = -hbar * wl(j) * 1/W * V * (1/(exp((hbar * wl(j)/(kb * T(i)))) - 1))
864
                      1/(exp((hbar*w1(j)/(kb*Teq)))-1))*((q1(j)^2)/(2*pi^2*v1(j)))*dw; ...
865
                          % LA number for hot side
                      nt(i,j)=-hbar*w2(j)*1/W*V*(1/(exp((hbar*w2(j)/(kb*T(i))))-1)
866
                      1/(exp((hbar*w2(j)/(kb*Teq)))-1))*((q2(j)^2)/(2*pi^2*v2(j)))*dw*2; ...
867
                          % TA number for hot side
                 end
868
                 n(i,j)=nl(i,j)+nt(i,j); % LA+TA
869
870
                 P(i,j)=nl(i,j)/n(i,j); % Polarization
871
             end
             NN(i)=floor(sum(n(i,:)));
872
            M(i, 1) = n(i, 1);
873
874
             F(i, 1) = 0;
             for j=2:1:1000
875
                 M(i,j)=M(i,(j-1))+n(i,j); % summation of ith phonon numbers
876
                 F(i,j)=M(i,j)/NN(i);% determine which frequency intervel
877
878
             end
879
        end
    %%%%%%%%%%% Phonon-electron scattering process
880
        u1=u1+1 % iteration
881
        dd(u1)=0; % number of particles under phonon-electron scattering
882
        for j=1:N
883
             for i=1:40
884
                 if (x1(j) > (i-1) * l \&\& x1(j) < (i) * l)
885
                      yy(j)=1.76*9.25/T(i);
886
                      E0=3.71e8; % constant
887
                      xm(j)=3.83; % coefficient for reduced frequency
888
889
                      if (mt(j)==0) % LA phonon
                           xf(j) = hbar \cdot f(j) / (kb \cdot T(i));
890
891
                           taol(j) = (2 \times E0 \times xf(j) \times T(i) \times exp(-yy(j)) \times (1 - xf(j) / (8 \times xm(j)))).^(-1);
892
                          taot(j)=0;
893
                      else % TA phonon
```

894	<pre>xf(j)=hbar*f(j)/(kb*T(i));</pre>
895	taot(j)=(2*E0*xf(j)*T(i)*exp(-yy(j))*(1-xf(j)/(8*xm(j)))).^(-1);
896	taol(j)=0;
897	end
898	<pre>tao(j)=taol(j)+taot(j); % Relaxation time</pre>
899	<pre>Ppe(j)=1-exp(-dt/tao(j)); % scattering probability</pre>
900	R=rand; % Random number
901	if (R <ppe(j))< td=""></ppe(j))<>
902	<pre>dd(u1)=dd(u1)+1; % particle is scattered, reassignment</pre>
903	R1=rand; % random number
904	al=1; % frequeny invervel No. 1
905	b1=1000; % frequeny invervel No. 1000
906	<pre>while((R1-F(i,a1))*(F(i,a1+1)-R1)&lt;0)</pre>
907	<pre>mm=round((al+b1)/2); % get integer</pre>
908	if (abs(R1-F(i,a1)) <abs(r1-f(i,b1))) %<="" td=""></abs(r1-f(i,b1)))>
	bisection method to determine which interval
909	if (R1>F(i,mm))
910	al=mm;
911	else
912	bl=mm;
913	end
914	else
915	<pre>if (R1<f(i,mm))< pre=""></f(i,mm))<></pre>
916	bl=mm;
917	else
918	al=mm;
919	end
920	end
921	end
922	R2=rand; %random number
923	R3=rand; %random number
924	f(j) = w1(a1) + (2 + R2 - 1) + dw/2; % reset the frequency
925	<pre>if (R3<p(i,a1)) %="" (la)<="" polarization="" pre=""></p(i,a1))></pre>

926	$ml(j) = (-vs(1) + sqrt(vs(1)^2 + 4 * c(1) * f(j))) / (2 * c(1));$	
	% LA wavevector	
927	<pre>mt(j)=0; % TA wavevector=0</pre>	
928	<pre>vl(j)=vs(1)+2*c(1)*ml(j); % LA magnitude group</pre>	
	velocity	
929	<pre>vt(j)=0; % TA velocity=0</pre>	
930	else % Polarization (TA)	
931	$mt(j) = (-vs(2) + sqrt(vs(2)^{2}+4*c(2)*f(j))) / (2*c(2));$	
	% TA wavevector (assume isotropy)	
932	<pre>ml(j)=0; % LA wavevector=0</pre>	
933	<pre>vt(j)=vs(2)+2*c(2)*mt(j); % TA magnitude group</pre>	
	velocity	
934	vl(j)=0; % LA velocity=0	
935	end	
936	v(j)=(vt(j)+vl(j)); % total velocity	
937	<pre>m(j)=mt(j)+ml(j); % total wave number</pre>	
938	R1=rand; % random number	
939	R2=rand; % random number	
940	K1=2*R1-1; % angle 1 psi	
941	K2=2*pi*R2; % angle 2 cos(theta)	
942	<pre>vg(j,2)=v(j)*sqrt(1-K1^2)*cos(K2); % direction of</pre>	
	velocity sin(theta)*cos(psi)	
943	<pre>vg(j,3)=v(j)*sqrt(1-K1^2)*sin(K2); % direction of</pre>	
	velocity sin(theta)*sin(psi)	
944	<pre>vg(j,1)=v(j)*K1; % direction of velocity cos(theta)</pre>	
945	end	
946	end	
947	end	
948	end	
949	<pre>pe(u1)=dd(u1)/N; % ratio of scattered particles</pre>	
950	8%%%% phonon dislocation scattering	
951	A0=0.8639; % constant for screw dislocations	
952	Nd=4.76e13; % dislocation density	

```
953
        gama=1.4; % Gruneissen constant
        b0=2.85e-10; % Burger's vector
954
        md(u1)=0; % number of particles under phonon-dislocation scattering
955
956
        for j=1:N
            for i=1:40
957
                 if (x1(j) > (i-1) * l \&\& x1(j) < (i) * l)
958
959
                     tau_d(j) = (A0*gama^2*b0^2*Nd*f(j))^(-1); % relacation time
                     Pd(j)=1-exp(-dt/tau_d(j)); % scattering probability
960
                     R=rand; % random number
961
                     if (R<Pd(j))</pre>
962
                         md(u1)=md(u1)+1; % particle is scattered, only ...
963
                             change velocity direction
                         R1=rand; % random number
964
                          R2=rand; % random number
965
                         K1=2*R1-1; % angle 2
966
                          K2=2*pi*R2; % angle 1
967
                          vg(j,2)=v(j)*sqrt(1-K1^2)*cos(K2); % direction of ...
968
                             velocity sin?*cos?
                          vg(j,3)=v(j)*sqrt(1-K1^2)*sin(K2); % direction of ...
969
                             velocity sin?*sin?
                          vg(j,1)=v(j)*K1; % direction of velocity sin?
970
971
                     end
972
                 end
            end
973
974
        end
   %%%%%% initialized sign of particles
975
        for i=1:40
976
            Np(i,u1)=0;
977
978
            Nn(i, u1) = 0;
979
        end
        for j=1:N
980
981
            for i=1:40
982
                 if (x1(j) > (i-1) * l \&\& x1(j) < i * l)
```

```
983
                     if s(j)==1
                         Np(i,u1)=Np(i,u1)+1; % number of positive particles
984
                     else
985
986
                         Nn(i,u1)=Nn(i,u1)+1; % number of positive particles
987
                     end
988
                 end
989
            end
990
        end
    %%%%%%%%%% calculate the heat flux in each cell at each time step
991
        for i=1:40
992
993
            qqq(i,u1)=0;
994
        end
        for j=1:N
995
            for i=2:39
996
                 if (x1(j)>(i-1)*l && x1(j)<i*l)
997
                     qqq(i,u1)=qqq(i,u1)+vg(j,1)*(s(j)*W)/V; % total heat flux
998
999
                 end
1000
            end
1001
        end
        qq(u1)=sum(qqq(:,u1))/38; % average heat flux
1002
        kk(u1) =qq(u1) *37*1/(T(2)-T(39)); % k at each timestep
1003
        for i=1:40
1004
            Tml(ul,i)=T(i); % temperature of each cell at each timestep
1005
            Em1(u1,i)=Et(i); % energy of each cell at each timestep
1006
            Nm(u1,i)=NNN(i); % number of particles (energy) in each cell
1007
1008
        end
        Ett(u1)=sum(Et); % total energy at each timestep
1009
1010 end
1011
1012 %%% Calculate Standard Deviation
1013 STD=0;
1014 for i=1:40
1015
        STD=STD+(T(i)-yy(i))^2;
```

```
1016 end
1017 STD=sqrt(STD/40);
1018 kkk=0;
1019 for j=(u1-100):1:u1
        kkk=kkk+kk(j); % summation of k for the last 100 timesteps
1020
1021 end
1022 kkk=kkk/101 % display average k
1023 Teq % display temperature
1024 plot(T) % plot temperature profle at the last step
1025 tim = toc;
1026 fprintf('operation time %15.5f.\n',tim); % calculate the operation time
1027 %%%%% Plat transient temperature profile
1028 i=1:40;
1029 plot(i,Tm1(1,i),'r',i,Tm1(100,i),'g',i,Tm1(500,i),'b',i,Tm1(1000,i),'m',i,Tm1(2000,i),'
1030 xlabel('Cell Number', 'FontSize', 12, 'FontName', 'Times New Roman');
1031 ylabel('Temperature/K', 'FontSize', 12, 'FontName', 'Times New Roman');
1032 legend('1*dt','100*dt','1000*dt','2000*dt','3000*dt')
1033 title('Energy-Based Variance-Reduced Method 50 nm/cell with Hao ...
       scattering')
```

## A.1.1 Child functions

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function zl=ff(T) %%%Function of zl
4 global V hbar kb dw wl w2 ql q2 vl v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*wl(i)*V*(1/(exp((hbar*wl(i)/(kb*T)))-1)-
9 1/(exp((hbar*wl(i)/(kb*Teq)))-1))*((ql(i).^2)/(2*pi^2*vl(i)))*dw;
```

```
10 %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
14 ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
15 1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
16 %%% Energy of TA polarization
17 end
18
19 z1=ztol-Et(1); %%%Function of z1 is related to Et.1
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z2=ff(T) %%%Function of z2
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar + w1(i) + V + (1/(exp((hbar + w1(i)/(kb + T))) - 1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
       %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z2=ztol-Et(2); %%%Function of z2 is related to Et.2
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z3=ff(T) %%%Function of z3
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+q(1)*hbar*wl(i)*V*(1/(exp((hbar*wl(i)/(kb*T)))-1)-
8
9
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
      %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
      ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
16
      %%%% Energy of TA polarization
17 end
18
19 z3=ztol-Et(3); %%%Function of z3 is related to Et.3
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z4=ff(T) %%%Function of z4
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
```

```
ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
9
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
       %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(\exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z4=ztol-Et(4); %%%Function of z4 is related to Et.4
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z5=ff(T) %%%Function of z5
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
      ztol=ztol+q(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
      %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
```

```
18
19 z5=ztol-Et(5); %%%Function of z5 is related to Et.5
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z6=ff(T) %%%Function of z6
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
14
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z6=ztol-Et(6); %%%Function of z6 is related to Et.6
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z7=ff(T) %%%Function of z7
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
```

```
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
       ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
9
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
       %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(\exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z7=ztol-Et(7); %%%Function of z7 is related to Et.7
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z8=ff(T) %%%Function of z8
4 global V hbar kb dw w1 w2 g1 g2 v1 v2 g Teg; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
9
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
       %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+q(2) + hbar+w2(i) + V + (1/(exp((hbar+w2(i)/(kb+T)))-1) - (hbar+w2(i)/(kb+T))) - 1)
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
```

```
16 %%%% Energy of TA polarization
17 end
18
19 z8=ztol-Et(8); %%%Function of z8 is related to Et.8
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z9=ff(T) %%%Function of z9
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8
      ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) -
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
      %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
      ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z9=ztol-Et(9); %%%Function of z9 is related to Et.9
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z10=ff(T) %%%Function of z10
```

```
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z10=ztol-Et(10); %%%Function of z1 is related to Et.10
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z11=ff(T) %%%Function of z11
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
      %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
```

```
14 ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
15 1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
16 %%% Energy of TA polarization
17 end
18
19 z11=ztol-Et(11); %%%Function of z11 is related to Et.11
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z12=ff(T) %%%Function of z12
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+q(2) + hbar+w2(i) + V + (1/(exp((hbar+w2(i)/(kb+T)))-1) - 1)
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z12=ztol-Et(12); %%%Function of z12 is related to Et.12
20 end
```

1 %%%%%% Child functions to calculate T using Newton-Raphson methos

```
2
3 function z13=ff(T) %%%Function of z13
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
8
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
      %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
      ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z13=ztol-Et(13); %%%Function of z13 is related to Et.13
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z14=ff(T) %%%Function of z14
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%% Energy of LA polarization
11 end
```

```
12
13 for i=1:1000
14     ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
15      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
16     %%% Energy of TA polarization
17 end
18
19 z14=ztol-Et(14); %%%Function of z14 is related to Et.14
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z15=ff(T) %%%Function of z15
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8
       ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
       %%%% Energy of LA polarization
10
11 end
12
13 for i=1:1000
       ztol=ztol+q(2) + hbar+w2(i) + V + (1/(exp((hbar+w2(i)/(kb+T)))-1) - 1)
14
15
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
       %%%% Energy of TA polarization
16
17 end
18
19 z15=ztol-Et(15); %%%Function of z15 is related to Et.15
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z16=ff(T) %%%Function of z16
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z16=ztol-Et(16); %%%Function of z16 is related to Et.16
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z17=ff(T) %%%Function of z17
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
11 end
12
13 for i=1:1000
14      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
15      l/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
16      %%% Energy of TA polarization
17 end
18
19 z17=ztol-Et(17); %%%Function of z17 is related to Et.17
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z18=ff(T) %%%Function of z18
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z18=ztol-Et(18); %%%Function of z18 is related to Et.18
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z19=ff(T) %%%Function of z19
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z19=ztol-Et(19); %%%Function of z19 is related to Et.19
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z20=ff(T) %%%Function of z20
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z21=ff(T) %%%Function of z21
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z21=ztol-Et(21); %%%Function of z1 is related to Et.21
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z22=ff(T) %%%Function of z22
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z22=ztol-Et(22); %%%Function of z22 is related to Et.22
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z3=ff(T) %%%Function of z23
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z24=ff(T) %%%Function of z24
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z24=ztol-Et(24); %%%Function of z24 is related to Eq.24
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z25=ff(T) %%%Function of z25
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z25=ztol-Et(25); %%%Function of z25 is related to Eq.25
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z26=ff(T) %%%Function of z26
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z27=ff(T) %%%Function of z27
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z27=ztol-Et(27); %%%Function of z1 is related to Eq.27
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z28=ff(T) %%%Function of z28
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z28=ztol-Et(28); %%%Function of z28 is related to Eq.28
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z29=ff(T) %%%Function of z29
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z30=ff(T) %%%Function of z1
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z30=ztol-Et(30); %%%Function of z30 is related to Eq.30
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z31=ff(T) %%%Function of z31
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z31=ztol-Et(31); %%%Function of z31 is related to Eq.31
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z32=ff(T) %%%Function of z32
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z33=ff(T) %%%Function of z33
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z33=ztol-Et(33); %%%Function of z33 is related to Eq.33
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z34=ff(T) %%%Function of z34
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z34=ztol-Et(34); %%%Function of z34 is related to Eq.34
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z35=ff(T) %%%Function of z35
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z36=ff(T) %%%Function of z36
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z36=ztol-Et(36); %%%Function of z1 is related to Eq.36
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z37=ff(T) %%%Function of z37
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z37=ztol-Et(37); %%%Function of z1 is related to Eq.37
20 end
```

```
1 %%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z38=ff(T) %%%Function of z38
4 global V hbar kb dw w1 w2 q1 q2 v1 v2 g Teq; %%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
8 ztol=ztol+g(1)*hbar*w1(i)*V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
9 1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
10 %%%% Energy of LA polarization
```

```
11 end
12
13 for i=1:1000
14      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
15      l/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
16      %%% Energy of TA polarization
17 end
18
19 z38=ztol-Et(38); %%%Function of z1 is related to Eq.38
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z39=ff(T) %%%Function of z39
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial energy is set to 0
7 for i=1:1000
       ztol=ztol+q(1) + hbar+w1(i) + V + (1/(exp((hbar+w1(i)/(kb+T)))-1) - 1)
8
       1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
       %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
       ztol=ztol+g(2) *hbar*w2(i) *V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
       1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
       %%%% Energy of TA polarization
16
17 end
18
19 z39=ztol-Et(39); %%%Function of z1 is related to Eq.39
20 end
```

```
1 %%%%%% Child functions to calculate T using Newton-Raphson methos
2
3 function z40=ff(T) %%%Function of z40
4 global V hbar kb dw wl w2 q1 q2 v1 v2 g Teq; %%%%% Global variables
5 global Et; %%%% Global variable of total energy
6 ztol=0; %%%% Initial enengy is set to 0
7 for i=1:1000
      ztol=ztol+g(1) *hbar*w1(i) *V*(1/(exp((hbar*w1(i)/(kb*T)))-1)-
8
      1/(exp((hbar*w1(i)/(kb*Teq)))-1))*((q1(i).^2)/(2*pi^2*v1(i)))*dw;
9
10
      %%%% Energy of LA polarization
11 end
12
13 for i=1:1000
      ztol=ztol+g(2)*hbar*w2(i)*V*(1/(exp((hbar*w2(i)/(kb*T)))-1)-
14
      1/(exp((hbar*w2(i)/(kb*Teq)))-1))*((q2(i).^2)/(2*pi^2*v2(i)))*dw;
15
      %%%% Energy of TA polarization
16
17 end
18
19 z40=ztol-Et(40); %%%Function of z1 is related to Eq.40
20 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function zldot=ff(T) %%%Function of zldot
3 global V hbar kb dw wl w2 ql q2 vl v2 g; %%%% Global variables
4 ztoldot=0; %%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(l)*V*hbar/(2*pi^2)*dw*(wl(i)*ql(i)^2/vl(i))*(l/(exp
7 (hbar*wl(i)/(kb*T))-l))^2*exp(hbar*wl(i)/(kb*T))*hbar*wl(i)/(kb*T.^2);
8 %%% LA polarization
9 end
10
```
```
11 for i=1:1000
12     ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13     (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14     %%% TA polarization
15 end
16 zldot=ztoldot; %%%Function of zldot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z2dot=ff(T) %%%Function of z2dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z2dot=ztoldot; %%%Function of z2dot is related to the derivative
17 end
```

```
    %%%%% Child dot functions to calculate T using Newton-Raphson methos
    function z3dot=ff(T) %%%Function of z3dot
    global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
    ztoldot=0; %%%% Initial zdot is set to 0
```

```
167
```

```
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
       %%%% LA polarization
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z3dot=ztoldot; %%%Function of z3dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z4dot=ff(T) %%%Function of z4dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
7
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
12
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z4dot=ztoldot; %%%Function of z4dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z5dot=ff(T) %%%Function of z5dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
13
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
      %%%% TA polarization
14
15 end
16 z5dot=ztoldot; %%%Function of z5dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z6dot=ff(T) %%%Function of z6dot
3 global V hbar kb dw wl w2 gl g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^{2}*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+q(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
```

```
14 %%%% TA polarization
15 end
16 z6dot=ztoldot; %%%Function of z6dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z7dot=ff(T) %%%Function of z7dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^{2}*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z7dot=ztoldot; %%%Function of z7dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z8dot=ff(T) %%%Function of z8dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(w1(i)*q1(i)^2/v1(i))*(1/(exp
7 (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
```

```
8 %%%% LA polarization
9 end
10
11 for i=1:1000
12 ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13 (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14 %%%% TA polarization
15 end
16 z8dot=ztoldot; %%%Function of z8dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z9dot=ff(T) %%%Function of z9dot
3 global V hbar kb dw wl w2 gl g2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
7
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z9dot=ztoldot; %%%Function of z9dot is related to the derivative
17 end
```

1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos

```
2 function z10dot=ff(T) %%%Function of z10dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
      %%%% LA polarization
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z10dot=ztoldot; %%%Function of z10dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z1ldot=ff(T) %%%Function of z1dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
```

```
15 end
16 zlldot=ztoldot; %%%Function of zlldot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z12dot=ff(T) %%%Function of z12dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw* (w2(i) *q2(i) ^2/v2(i)) * (1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
14
      %%%% TA polarization
15 end
16 z12dot=ztoldot; %%%Function of z12dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z13dot=ff(T) %%%Function of z13dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(w1(i)*q1(i)^2/v1(i))*(1/(exp
7 (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
8 %%% LA polarization
```

```
9 end
10
11 for i=1:1000
12     ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13     (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14     %%%% TA polarization
15 end
16 z13dot=ztoldot; %%%Function of z13dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z14dot=ff(T) %%%Function of z14dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
       %%%% LA polarization
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+q(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z14dot=ztoldot; %%%Function of z14dot is related to the derivative
17 end
```

%%%%%% Child dot functions to calculate T using Newton-Raphson methos
 function z15dot=ff(T) %%%Function of z15dot

```
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z15dot=ztoldot; %%%Function of z15dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z16dot=ff(T) %%%Function of z16dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
```

```
16 z16dot=ztoldot; %%%Function of z16dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z17dot=ff(T) %%%Function of z17dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z17dot=ztoldot; %%%Function of z17dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function zl8dot=ff(T) %%%Function of zl8dot
3 global V hbar kb dw wl w2 ql q2 vl v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(wl(i)*ql(i)^2/vl(i))*(1/(exp
7 (hbar*wl(i)/(kb*T))-1))^2*exp(hbar*wl(i)/(kb*T))*hbar*wl(i)/(kb*T.^2);
8 %%% LA polarization
9 end
```

```
10
11 for i=1:1000
12     ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13     (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14     %%% TA polarization
15 end
16 z18dot=ztoldot; %%%Function of z18dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z19dot=ff(T) %%%Function of z19dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z19dot=ztoldot; %%%Function of z19dot is related to the derivative
17 end
```

```
    %%%%% Child dot functions to calculate T using Newton-Raphson methos
    function z20dot=ff(T) %%%Function of z20dot
    global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
```

```
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
7
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
13
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
       %%%% TA polarization
14
15 end
16 z20dot=ztoldot; %%%Function of z20dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z21dot=ff(T) %%%Function of z21dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z21dot=ztoldot; %%%Function of z21dot is related to the derivative
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z22dot=ff(T) %%%Function of z22dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
12
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z22dot=ztoldot; %%%Function of z22dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z23dot=ff(T) %%%Function of z23dot
3 global V hbar kb dw wl w2 ql q2 vl v2 g; %%%% Global variables
4 ztoldot=0; %%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(wl(i)*ql(i)^2/vl(i))*(1/(exp
7 (hbar*wl(i)/(kb*T))-1))^2*exp(hbar*wl(i)/(kb*T))*hbar*wl(i)/(kb*T.^2);
8 %%% LA polarization
9 end
10
```

```
11 for i=1:1000
12 ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13 (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14 %%%% TA polarization
15 end
16 z23dot=ztoldot; %%%Function of z23dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z24dot=ff(T) %%%Function of z24dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z24dot=ztoldot; %%%Function of z24dot is related to the derivative
17 end
```

```
    %%%%% Child dot functions to calculate T using Newton-Raphson methos
    function z25dot=ff(T) %%%Function of z25dot
    global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
    ztoldot=0; %%%% Initial zdot is set to 0
```

```
5 for i=1:1000
6
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
       %%%% LA polarization
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z25dot=ztoldot; %%%Function of z25dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z26dot=ff(T) %%%Function of z26dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
7
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
12
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z26dot=ztoldot; %%%Function of z26dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z27dot=ff(T) %%%Function of z27dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
13
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
      %%%% TA polarization
14
15 end
16 z27dot=ztoldot; %%%Function of z27dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z28dot=ff(T) %%%Function of z28dot
3 global V hbar kb dw wl w2 gl g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+q(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
```

```
14 %%%% TA polarization
15 end
16 z28dot=ztoldot; %%%Function of z28dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z29dot=ff(T) %%%Function of z29dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z29dot=ztoldot; %%%Function of z29dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z30dot=ff(T) %%%Function of z30dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(w1(i)*q1(i)^2/v1(i))*(1/(exp
7 (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
```

```
8 %%%% LA polarization
9 end
10
11 for i=1:1000
12 ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13 (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14 %%%% TA polarization
15 end
16 z30dot=ztoldot; %%%Function of z30dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z3ldot=ff(T) %%%Function of z3ldot
3 global V hbar kb dw wl w2 gl g2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
7
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z31dot=ztoldot; %%%Function of z31dot is related to the derivative
17 end
```

1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos

```
2 function z32dot=ff(T) %%%Function of z32dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
      %%%% LA polarization
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z32dot=ztoldot; %%%Function of z32dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z33dot=ff(T) %%%Function of z33dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
```

```
15 end
16 z33dot=ztoldot; %%%Function of z33dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z34dot=ff(T) %%%Function of z34dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
14
      %%%% TA polarization
15 end
16 z34dot=ztoldot; %%%Function of z34dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z35dot=ff(T) %%%Function of z35dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(w1(i)*q1(i)^2/v1(i))*(1/(exp
7 (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
8 %%% LA polarization
```

```
9 end
10
11 for i=1:1000
12     ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13     (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14     %%%% TA polarization
15 end
16 z35dot=ztoldot; %%%Function of z35dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z36dot=ff(T) %%%Function of z36dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
8
       %%%% LA polarization
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+q(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z36dot=ztoldot; %%%Function of z36dot is related to the derivative
17 end
```

%%%%% Child dot functions to calculate T using Newton-Raphson methos
 function z37dot=ff(T) %%%Function of z37dot

```
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw* (w1(i) *q1(i) ^2/v1(i)) * (1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
16 z37dot=ztoldot; %%%Function of z37dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z38dot=ff(T) %%%Function of z38dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
       ztoldot=ztoldot+q(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
       %%%% LA polarization
8
9 end
10
11 for i=1:1000
       ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
       %%%% TA polarization
14
15 end
```

```
16 z38dot=ztoldot; %%%Function of z38dot is related to the derivative
17 end
```

```
1 %%%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z39dot=ff(T) %%%Function of z39dot
3 global V hbar kb dw w1 w2 g1 g2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
      ztoldot=ztoldot+g(1) *V*hbar/(2*pi^2) *dw*(w1(i) *q1(i) ^2/v1(i)) *(1/(exp
6
       (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
7
      %%%% LA polarization
8
9 end
10
11 for i=1:1000
      ztoldot=ztoldot+g(2) *V*hbar/(2*pi^2) *dw*(w2(i) *q2(i) ^2/v2(i)) *(1/(exp
12
       (hbar*w2(i)/(kb*T))-1))^{2}*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
13
      %%%% TA polarization
14
15 end
16 z39dot=ztoldot; %%%Function of z39dot is related to the derivative
17 end
```

```
1 %%%%% Child dot functions to calculate T using Newton-Raphson methos
2 function z40dot=ff(T) %%%Function of z40dot
3 global V hbar kb dw w1 w2 q1 q2 v1 v2 g; %%%%% Global variables
4 ztoldot=0; %%%% Initial zdot is set to 0
5 for i=1:1000
6 ztoldot=ztoldot+g(1)*V*hbar/(2*pi^2)*dw*(w1(i)*q1(i)^2/v1(i))*(1/(exp
7 (hbar*w1(i)/(kb*T))-1))^2*exp(hbar*w1(i)/(kb*T))*hbar*w1(i)/(kb*T.^2);
8 %%% LA polarization
9 end
```

```
10
11 for i=1:1000
12     ztoldot=ztoldot+g(2)*V*hbar/(2*pi^2)*dw*(w2(i)*q2(i)^2/v2(i))*(1/(exp
13        (hbar*w2(i)/(kb*T))-1))^2*exp(hbar*w2(i)/(kb*T))*hbar*w2(i)/(kb*T.^2);
14      %%%% TA polarization
15 end
16 z40dot=ztoldot; %%%Function of z40dot is related to the derivative
17 end
```

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