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EFFECT OF DISLOCATIONS ON THE THERMAL CONDUCTIVITY OF SUPERCONDUCTING Nb

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Abstract

The thermal conductivity of Niobium (Nb) often experiences a local maximum (a phonon peak) at a temperature between 1.8 and 3 K. While the magnitude of the phonon peak has been shown to be related to the dislocation density and may be influenced by manufacturing processes, little has been discussed as to the temperature at which the peak occurs. In examining these phenomena, it has been determined that more explicit accounting of phonon–dislocation scattering in a popular model better represents the thermal conductivity at temperatures colder than 3 K. Scaled sensitivity coefficients show this term to have similar influence as the phonon-electron and phonon-boundary scattering terms. Results using the enhanced model also show an apparent threshold of dislocation density ($N_d < 10^{12}$ m^{-2}) below which there is little contribution to the thermal conductivity of Nb.

INTRODUCTION

Manufacturing superconducting radio frequency (SRF) cavities from large grain niobium (Nb) may reduce cost and improve the quality factor as compared with polycrystalline Nb [1]. Processing Nb to obtain the largest thermal conductivity possible is an important component of the improved performance. Even in the superconducting regime, small imperfections at the RF surface can cause local heating that leads to loss of performance. Large values of thermal conductivity can mitigate local temperature excursions and prevent cavity quench, thus improving cavity performance [2].

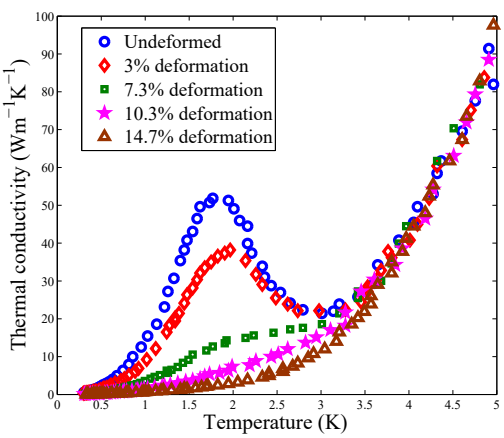

Figure 1: Thermal conductivity of sample K6 from Wasserbäch with different deformation, replotted from [3].

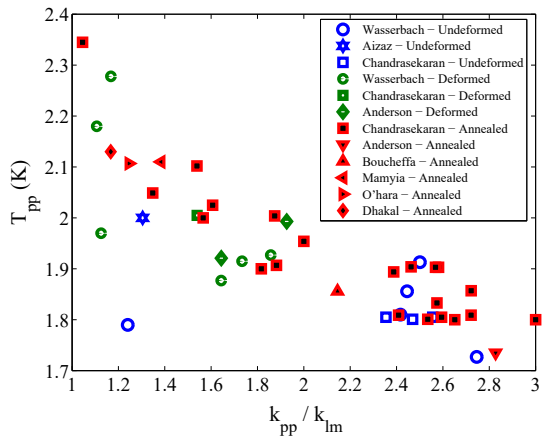

Figure 2: Measured phonon peak temperatures T_{pp} for undeformed, deformed and annealed specimens as a function of the ratio of the thermal conductivity at phonon peak k_{pp} to that at local minimum temperature k_{lm}.

Manufacturing SRF cavities from Nb sheets requires large deformations that increase dislocation density [4], which has been shown to reduce the thermal conductivity of superconducting large grain Nb [3, 5–9]. Wasserbäch measured the thermal conductivity of Nb after uniaxial straining of up to 22.2% [3]. An example of these data for a single specimen that has undergone increasing levels of deformation is replotted in Fig. 1, where the thermal conductivity k for temperatures colder than 3 K decreases with increasing deformation. Of particular note is the local maximum in k (*i.e.*, the phonon peak in conductivity k_{pp}) at approximately 2 K that diminishes with increasing deformation. Wasserbäch examined the effect of deformation on conduction [9] by using a relaxation time approximation [10, 11]. Phonon-electron scattering, which is a significant factor at the working temperatures of SRF cavities (about 2 K), 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 the local maximum phonon peak and the decrease in dislocation density. A phonon peak that has disappeared after deformation can be partially recovered with heat treatment of appropriate temperature and duration [12]. In the analysis of effect of deformation on conductivity, only k_{pp} was used to estimate the dislocation density. Koechlin and Bonin [13] used a simplified equation based on the Bardeen-Rickayzen-Tewordt (BRT) model [14] to fit the experimental data. This equation was reparameterized by Chandrasekaran [15] for analysis and

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to reduce uncertainties in estimating the thermal conductivity of Nb. However, neither of these equations considered phonon-dislocation scattering, which might be the reason that Koechlin and Bonin note discrepancies in fitting of k at temperatures lower than 2 K [13].

A little discussed observation that can be seen in Fig. 1 is the variation in temperatures at which k_{pp} occurs. As k_{pp} decreases, the temperature at this it occurs increases. Typically, the temperature of the phonon peak T_{pp} is cited as occurring at from $1.8 \leq T_{pp} \leq 2$ K. Figure 2 shows reported values of $1.72 \leq T_{pp} \leq 2.35$ K for a number of studies of large grain Nb [3, 6–8, 16–20]. This suggests that both k_{pp} and T_{pp} are functions of the dislocation density, and thus the deformation of Nb. This further suggests that phonon–dislocation scattering should be accounted for explicitly in the model of k. This addition, and some of its implications, is considered here.

ANALYSIS

Observations The results mentioned above and shown in Fig. 2 represent measurements from several studies for as received, deformed, and annealed specimens [3, 6–8, 16–20]. The temperature data are plotted as a function of the ratio k_{pp}/k_{lm}, where k_{lm} is the local minimum in conductivity, usually at about 3 K. Chandrasekaran [8] showed that k_{pp}/k_{lm} correlates inversely with dislocation density for a given specimen having undergone deformation or heat treatment. Although there is scatter in the data in Fig. 2, resulting from the sample processing history and experimental technique, 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 starts to dominate the thermal conductivity.

Figure 2 further shows that undeformed samples typically have colder T_{pp}, but greater k_{pp}. For deformations typically less than 3% strain, k_{pp} decreases and T_{pp} increases. For uniaxial straining greater than 3%, the phonon peak disappears in most samples because of phonon-dislocation scattering. High purity samples, however, maintain a phonon peak after more deformation (*e.g.*, the sample shown with RRR=1200 still has phonon peak after 4% uniaxial straining [3]). Therefore, deformation not only affects the value of thermal conductivity, but also has an apparent effect on T_{pp}. Heat treatment can reduce dislocation content [16] to partially or fully recover 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 the undeformed state.

Modeling The thermal conductivity of Nb can be modeled as consisting of electron transport of energy and phonon of energy. These two components are additive and can be expressed as

$$k = k_e + k_g \tag{1}$$

where k_e represents thermal energy transport by electrons and k_g represents the thermal energy transport due to phonons. In normally conducting metals, the phonon part is usually negligible due to scattering by normal electrons. However, in the superconducting regime, the formation of 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 [21]. Therefore, the phonon contribution to thermal conduction increases in significance in superconducting materials.

Koechlin and Bonin [13] modeled the thermal conductivity of superconducting Nb by parameterizing the BRT [14] expression according to Bardeen-Cooper-Schrieffer (BCS) theory [21] as

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

where $R(y)$ quantifies the condensation of normal conducting electrons into Cooper pairs [14], ρ is the residual resistivity ($\rho = \rho_{295K}/RRR$), RRR is the residual resistivity ratio, ρ_{295K} is the electrical resistivity at 295 K. $L = 2.45 \times 10^{-8}$ WK^{-2} is the Lorentz number, a is the coefficient of momentum exchange of electrons with the lattice, D refers to phonon electron scattering, B corresponds to phonon boundary scattering, and Λ is the phonon mean free path. The term y in $R(y)$ is defined as

$$y = \frac{\Delta(T)}{k_B T} = \frac{\Delta(T)}{k_B T_c}\frac{T_c}{T} \tag{3}$$

where $\Delta(T)$ is the superconducting energy gap, and k_B the Boltzmann constant. For $T/T_c < 0.6$, y can be approximated as $y = \alpha T_c/T$, $\alpha \approx 1.76$ in BCS theory, and ρ, a, B, and D are the four parameters that need to be determined. The two terms in k_e are due to electron-defect scattering and electron-phonon scattering, respectively, and the two terms in k_g are due to phonon-electron scattering and phonon boundary scattering, respectively.

As mentioned above, phonon-dislocation scattering should be included in the model, especially for Nb following deformation. Adding a dislocation term to Eq. (2) yields

$$k = R(y)\left[\frac{\rho}{LT} + aT^2\right]^{-1} + \left[\frac{1}{D\exp(y)T^2} + \frac{1}{B\Lambda T^3} + W_d\right]^{-1} \tag{4}$$

where the phonon dislocation scattering thermal resistance W_d is expressed following Klemens [11] for randomly distributed dislocations as

$$\frac{W_d T^2}{N_d} = \frac{0.038(\bar{\nu}h^2)b^2\gamma^2}{{k_B}^3} \tag{5}$$

where N_d is the dislocation density, γ is the Grüneisen constant, γ=1.4 for Nb, b is the Burgers vector, $\bar{\nu}$ is the average group velocity of Nb, h is the Planck constant and k_B is the

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Boltzmann constant. Then, for Nb, an expression for W_d may be written as

$$\frac{W_d T^2}{N_d} = 3.05 \times 10^{-15}\ \mathrm{m^3K^3/W} \qquad (6)$$

According to Wasserbäch [3], Bross analyzed the components of thermal resistance in copper for edge and screw dislocations. He obtained results to be 1.67 and 1.26 greater for edge and screw dislocations, respectively, than the values evaluated by Klemens [11].

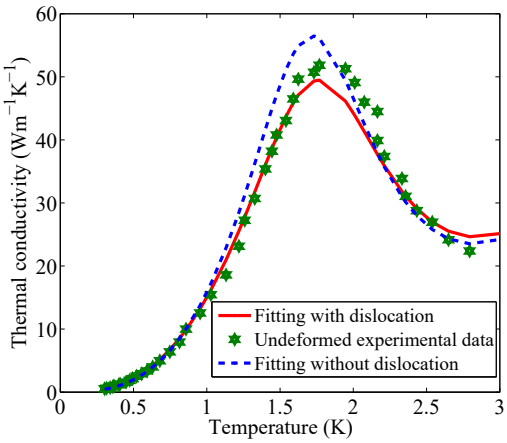

Figure 3: Comparison between fitting with or without dislocation term for undeformed sample from [3]. Calculated dislocation density is $N_d = 4.67 \times 10^{12}$ m^{-2}.

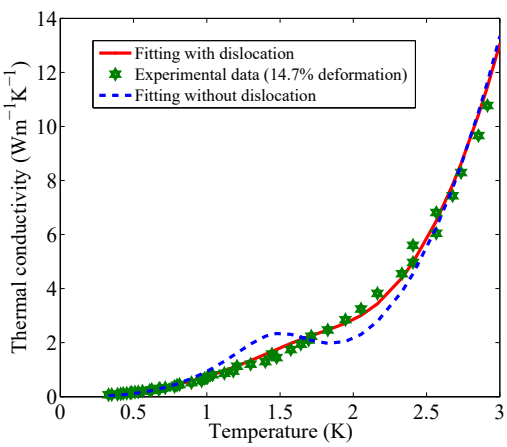

Figure 4: Comparison between fitting with or without dislocation term for deformed sample [3]. Calculated dislocation density is $N_d = 3.83 \times 10^{14}$ m^{-2}.

RESULTS AND DISCUSSION

Comparison of data from [3] with fitting of the model are shown in Fig. 3 for undeformed sample and Fig. 4 for the same sample after uniaxial straining of 14.7%. Fitting including the dislocation term improves comparison qualitatively and quantitively with the experimental results [3], especially for the Nb sample after deformation. This improvement in agreement suggests that the observation by Koechlin and Bonin of a deviation in the fitting of their low temperature data using Eq. (2) [13] 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.

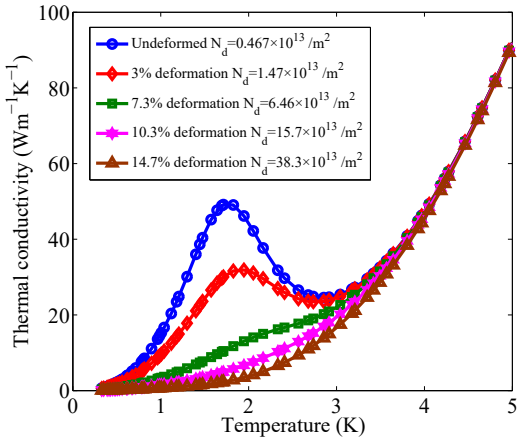

Figure 5: Thermal conductivity of sample K6 (an example of edge dislocation) [3] using curve fitting by considering the effect of dislocation.

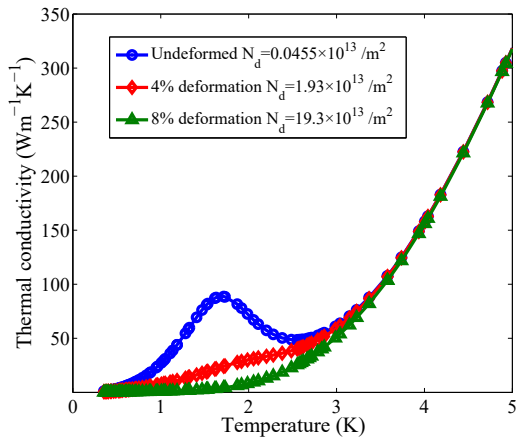

Figure 6: Thermal conductivity of sample H2 (an example of edge dislocation) [3] using curve fitting by considering the effect of dislocation.

Several other sets of data from Wasserbäch [3] were fit using the above method to examine the role of dislocation density and its effect on the thermal conductivity of Nb. These fits are shown in Fig. 5 for edge dislocations and Fig. 6 for screw dislocations. Wasserbäch [3] calculated the dislocation density for sample K6 to be about 10^{14} m^{-2} after the last deformation (14.7%), without including phonon-electron scattering. The dislocation density obtained here for the 14.7% deformation case is 3.83×10^{14} m^{-2}. It reaches

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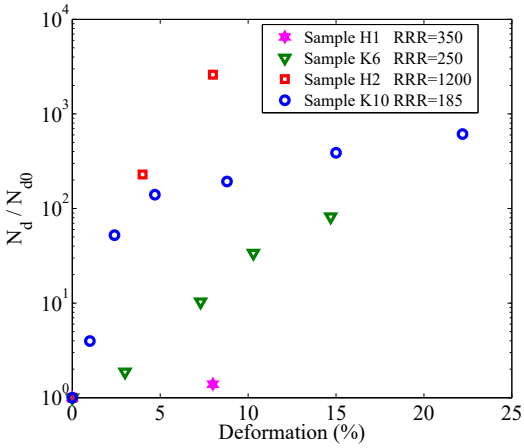

Figure 7: The ratio of deformed dislocation density to that before deformation as a function of deformation, determined by fitting of thermal conductivity. Sample H1, K6, H2, and K10 are deformed at temperature of 77 K, 195 K, 295 K, and 1470 K, respectively. H1 and K6 had screw dislocation, H2 and K10 had edge dislocation.

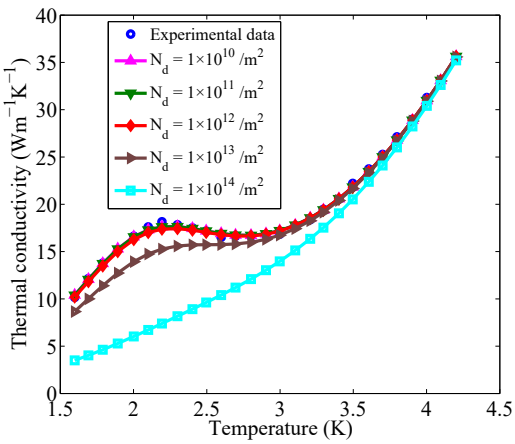

Figure 8: The effect of dislocation density on thermal conductivity of Nb at temperature between 1.5 K and 4.5 K.

10^{14} m^{-2} after 10% deformation. These are in reasonable agreement with value given by Wasserbäch [3].

The ratio of deformed dislocation density N_d to the dislocation density before deformation N_{d0} is plotted as a function of deformation (*i.e.*, uniaxial straining) and shown in Fig. 7 for sample of varying RRR from [3]. 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 6.11×10^{10} m^{-2}, however, the values for the other three samples are approximately 10^{12} m^{-2} with similar RRR values for 185, 250, and 350. It is also found

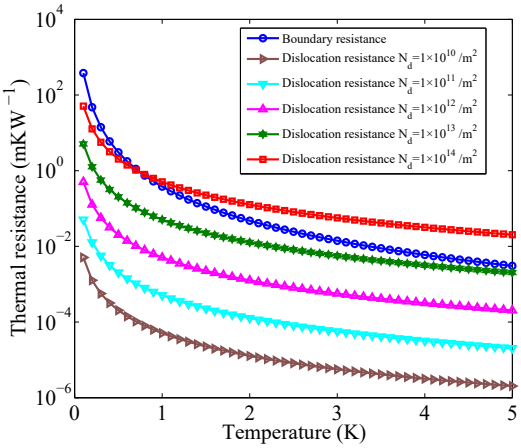

Figure 9: Comparison between phonon boundary thermal resistance and phonon dislocation resistance for different dislocation density.

that the phonon dislocation scattering only affects the thermal conductivity when it reaches a certain magnitude, *e.g.* 10^{12} m^{-2}, shown in Fig. 8 for measurements from [8]. The plots almost overlap with each other for $N_d < 10^{12}$ m^{-2}. The reason appears to be that phonon boundary scattering dominates heat conduction at low temperatures [22], and that the thermal resistance of phonon–dislocation scattering for $N_d < 10^{12}$ m^{-2} is much smaller than that of boundary scattering, as shown in Fig. 9.

CONCLUSIONS

For high performance for SRF applications, large values of thermal conductivity are desired for improved thermal stabilization. However, higher working temperature might yield significant savings in the total energy required for SRF cavity operation. 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.

Analysis of the thermal conductivity of superconducting Nb shows that in addition to k_{pp} decreasing after deformation, there is a shift increase in T_{pp}. The proposed model adds a phonon–dislocation scattering term that improves the accuracy of fits to experimental data, especially for deformed samples. The proposed model can also be used to infer the dislocation density from measurements of k. Results show that a threshold dislocation density exists below which there is no significant affect on the thermal conductivity. A dislocation density smaller than 10^{12} m^{-2} has little contribution to the thermal conductivity of superconducting Nb, because boundary scattering dominates at low temperatures.

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