Lattice thermal conductivity of Na

R. Fletcher
Physics Department, Queen's University, Kingston, Ontario, Canada K7L 3N6
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Measurements on the lattice thermal conductivity of Na in the liquid-^{4}He temperature range indicate that the phonon mean free path is independent of the phonon frequency and hence the temperature. The measured mean free path of 1.7 μm is believed to represent the scale of the martensitic transformation that occurs in Na below about 36 K.

It is known that the lattice thermal conductivity λ_g of K metal is anomalous in being typically an order of magnitude larger than theory predicts in the liquid-^{4}He temperature range (Fletcher,[1] referred to as I subsequently, gives references to earlier work). In contrast, λ_g for Rb is close to that expected.[2] The explanation of the difference is probably to be found in the degree to which each of these two metals approaches the free-electron model (i.e., with zero band gaps across the Brillouin-zone boundaries). When the metal is very free-electron-like, as with K, then the transverse phonons are almost uncoupled to the electrons and this leads to a very large λ_g. Rb seems to be sufficiently non-free-electron-like[3] that this no longer happens. Na has an even more spherical Fermi surface[4] than K, and on this basis the transverse phonons in Na should be even more decoupled from the electrons than in K. However, unlike K and Rb, Na undergoes a martensitic phase transformation[5] below about 36 K in which some fraction of the normal bcc phase is converted to hcp. The fraction that transforms has been estimated[5-7] to be anywhere in the region 7–50%, with large-grained samples probably being at the lower end of the range. Since we do not expect λ_g for Na to be limited by electronic scattering, we would expect it to be strongly influenced by the presence of the embedded hcp phase and the present experiments were performed to investigate the form that this takes. Within experimental error $\sim$0.2%, the two phases have identical atomic volumes[5,7] and so there is no reason to expect excessive strain to be introduced by the transformation, and hence one anticipates relatively few dislocations to be produced. From the nature of the transformation a high density of stacking faults is likely to be produced in the transformed region.[5] The fact that the residual resistivity ρ_0 of Na samples is independent of the fraction of hcp (Ref. 6) suggests that grain boundaries, dislocations, and stacking faults do not contribute significantly to ρ_0, though this does not necessarily imply a similar result for λ_g.

The Na was obtained from the MSA Corp., Callery, PA, and had a resistivity ratio $\rho(295\ \mathrm{K})/\rho(0\ \mathrm{K})$ of about 800. In previous studies, material with a much higher resistivity ratio has been obtained from the same source but in the present case this was the best of several vials that were examined. Rods of about 6 mm diam were extruded at about 40 °C–50 °C, and, using a specially designed clamp to hold the sample securely, the center 45 mm of each rod was trimmed to an approximately rectangular cross section of 1.5×6.0 mm^2. This helps to ensure that under the influence of an applied stress the strain is mainly confined to the thinned region. It also led to a higher reliability of the thermometers that were attached just outside this region. The samples were free-standing and subject to no appreciable strain on cooling.

The experimental details and method of analysis were similar to those described in I. Briefly, the method involves measuring the transverse magnetothermal resistivity of the sample and analyzing the contribution that varies as the square of the magnetic field B in terms of λ_g. We also observed a component linear in B that, when multiplied by L_0T (where L_0 is the Sommerfeld value of the Lorenz number), became independent of T and identical to within 5% with that observed in the transverse electrical magnetoresistivity. The values of λ_g extracted from the data are shown in Fig. 1. The sample was subsequently strained by 10% at $T\sim40$ K (with no observable necking) and then gave lower values of λ_g as shown in the

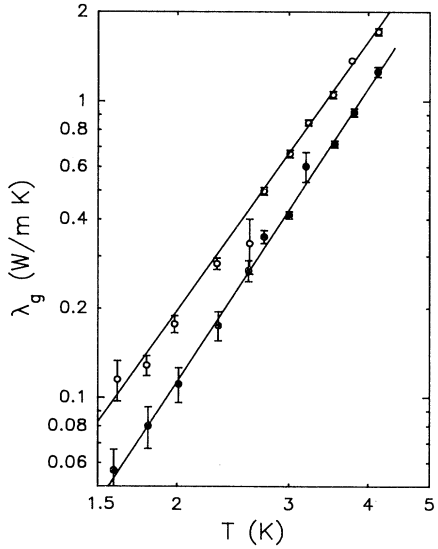

FIG. 1. The measured thermal conductivity of one of the Na samples as a function of temperature (on logarithmic scales). The open circles are for the as-cooled sample and the straight line through these data has a slope of 3.01±0.10. The solid circles are appropriate to the same sample after straining 10% at 40 K; the straight line through these data has a slope of 3.25±0.19.

same figure. It is interesting to note that at 4 K the lattice contributes only 0.1% to the total heat conductivity of the unstrained sample, and this fraction is considerably reduced at lower T. Another unstrained sample from the same batch exhibited the same λ_g within experimental error ($\sim 5\%$) but this sample broke when strained at 4.2 K (as did several others). The reproducibility between the two unstrained samples suggests that the same fraction of material transformed to hcp on cooling to 4.2 K.

Power-law fits of the form $\lambda_g \sim T^n$ gave $n = 3.01 \pm 0.10$ and 3.07 ± 0.12 for the two unstrained samples, and $n = 3.25 \pm 0.19$ for the single strained sample. The behavior in both unstrained and strained cases is consistent with $n = 3$, which implies a phonon mean free path Λ independent of phonon frequency. In such cases one finds λ_g to be given by[8]

$$\lambda_g = \frac{2\pi^2 k^4}{15} (T/\hbar)^3 \Lambda \langle 1/v^2 \rangle ,$$

where v is the velocity of the phonon branch and the average, denoted by $\langle\ \rangle$, is to be taken over the three polarizations and all directions. Using the elastic constant data of Diederich and Trivisonno[9] at 78 K, and averaging $\langle 1/v^2 \rangle$ over the [100], [110], and [111] directions, Λ is found to be about 1.7 μm for the unstrained sample and about 30% lower for the strained sample. The only scattering sources that can lead to a constant Λ are of large scale compared to the phonon wavelength such as the surfaces of the sample itself, which are obviously not relevant here, or much more likely, the embedded hcp phase. The source of the scattering seems likely to be the crystallite boundaries introduced by the transformation, though it is possible that the stacking faults within the hcp phase also contribute. Although the densities of the bcc and hcp phases are identical,[5,7] the phonon velocities will presumably be quite different and lead to a strong acoustic impedance mismatch.[10] A similar effect has been demonstrated in polycrystalline Al_2O_3, BeO, and graphite,[11] where the measured Λ were found to correspond to the crystallite dimensions. Before cooling, the grain size in the Na was ~ 1 mm, which is much too large to be relevant.

Stedman,[7] in a neutron-scattering study of the phonons in the bcc phase of Na at temperatures above and below the martensitic phase transition, also suggested that the transformation occurs on a fine scale. He found that the integrated intensity of the phonon peaks was reduced by a factor of 0.3 when the hcp phase was present and suggested that this might be viewed as an extreme case of a mosaic crystal where the individual regions are not coherently connected as far as phonons are concerned. This led Stedman to suggest a scale length of perhaps 100 atom spacings (~ 43 nm) for the transformation, which is about a factor of 40 smaller than the present estimate.

The observed temperature dependence of λ_g appears to eliminate the possibility of scattering by electrons and static dislocations which should both lead to[10] $\lambda_g = \alpha T^2$. In the case of electron scattering, we can use the analysis described in Ref. 2 (based on the measurement of the ideal electronic thermal resistivity that has been performed on these samples). This yields $\alpha = 0.06$ $\mathrm{W\,m^{-1}\,K^{-3}}$ or $\lambda_g \sim 1.0$ $\mathrm{W\,m^{-1}\,K^{-1}}$ at 4 K. This is roughly the same as experimentally observed but the incorrect power law shows that this must be coincidental. As mentioned earlier, a more realistic α is at least an order of magnitude larger assuming that Na and K behave similarly. An estimate is also possible for dislocation scattering. If the scattering of phonons by a single dislocation is taken to be similar in K and Na, then α is found to depend only on $\langle 1/v \rangle$ (see I for details). This factor is about 3 times smaller for Na than for K. If the K data are examined[1] at a strain of 2–3 % (since the atomic volumes of the hcp and bcc phases of Na are the same, it is difficult to see why the strain due to the transformation should be large), one finds that such a strain acting alone would lead to $\lambda_g \sim 30$ $\mathrm{W\,m^{-1}\,K^{-1}}$ at 4 K, and so for Na we would expect $\lambda_g \sim 10$ $\mathrm{W\,m^{-1}\,K^{-1}}$ at the same temperature. Again, this is an order of magnitude larger than the observed λ_g and does not appear to be important. Dislocation scattering will be stronger in the strained Na sample but the temperature dependence of λ_g suggests that even here it is not significant in the data. In principle, scattering by vibrating dislocations could give $\lambda_g \sim T^3$, but since no sign of this mechanism was observed for K (Ref. 1) it is assumed to be also absent here.

Templeton[12] has suggested that Rb might also undergo a martensitic phase transformation at low temperature, but the contrasting behavior that we see for Na and Rb (Ref. 2) suggests that this has not happened in our Rb samples down to at least 1.2 K.

To summarize, the observed cubic temperature dependence of λ_g that we observe for Na is characteristic of phonon scattering by large-scale imperfections. We associate the mean free path $\Lambda \sim 1.7$ μm that is observed with the scale of the martensitic transformation in Na. It would be of interest to correlate this scale with the fraction of hcp phase produced; in principle, the latter can be found from a measurement of the ideal resistivity,[6] but this was not possible in the present study.

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[1] R. Fletcher, Phys. Rev. B **36**, 3042 (1987).
[2] R. Fletcher and I. B. Verma, Phys. Rev. B **36**, 9482 (1987).
[3] D. Schoenberg and P. J. Stiles, Proc. R. Soc. London, Ser. A **281**, 62 (1964).
[4] M. J. G. Lee, Proc. R. Soc. London, Ser. A **295**, 440 (1966).
[5] C. S. Barrett, Acta Crystallogr. **9**, 671 (1955).
[6] J. S. Dugdale and D. Gugan, Proc. R. Soc. London, Ser. A **254**, 184 (1960).
[7] R. Stedman, J. Phys. F **6**, 2239 (1976).
[8] J. Callaway, Phys. Rev. **113**, 1046 (1959).
[9] M. E. Diederich and J. Trivisonno, J. Phys. Chem. Solids **27**, 637 (1966).
[10] J. M. Ziman, *Electroncs and Phonons* (Oxford University Press, London, 1960).
[11] R. Berman, Proc. Phys. Soc. London **65**, 1029 (1952).
[12] I. M. Templeton, J. Phys. F **12**, L121 (1982).