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THERMAL CONDUCTIVITY AND KAPITZA RESISTANCE IN THE NORMAL AND SUPERCONDUCTING STATES*

I. Introduction

There are two processes by which heat may be transported through a solid. One of the processes arises due to the strong coupling between the atoms in a crystal lattice. Thus thermal conduction can take place by means of the lattice vibrations or phonons. These can be considered as longitudinal and transverse waves travelling through the material. This is the mechanism of heat transport in dielectrics and, as we shall see, the dominant mechanism in superconductors.

The second process of heat conduction is by "free" electrons. In fairly pure metals, this is the dominant mechanism. As we shall see, this is why there is an intimate relationship between thermal conductivity and electrical conductivity in normal metals.

Though our main interest will be in low-temperature thermal conductivity, the basic ideas which will be developed are applicable over the whole temperature range. Thus we shall also briefly look at high and intermediate temperatures. Kapitza resistance will also be considered, as an example of general thermal boundary resistance between two media which becomes particularly manifest at low temperatures.

The derivations that will be presented are short, simple, and not rigorous. There isn't enough time to present the rigorous derivations. A lot of things will

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*These notes represent the subject matter covered in an informal Superconducting Journal Club seminar presented by Mario Rabinowitz.

be swept under the rug. These will not be pointed out as such to avoid confusion at this level, since the topic is not of great familiarity to most of the people here. I would be glad to personally discuss the finer points with those of you who are interested. I hope to get across the basic concepts and their broad scope of applicability. What the heuristic derivations lack in rigor, I hope they'll make up for in vigor to give you an insight into the physics behind the relevant equations.

Although most of the derivations and interpretations are my own (I take the responsibility for their incompleteness and the errors in them), I have relied on several standard texts for general guidance. I recommend them to those who wish to probe deeper into the subject. These texts are: Rosenberg's Low Temperature Solid State Physics, Ziman's Electrons and Phonons, Dekker's Solid State Physics, and Kittel's Introduction to Solid State Physics.

II. The Wiedemann-Franz Law

Let's start by quickly looking at electrical conductivity with which we're familiar, and then look at the contribution to thermal conductivity due to electrons by analogy. We'll then relate the two conductivities.

A. Electrical Conductivity

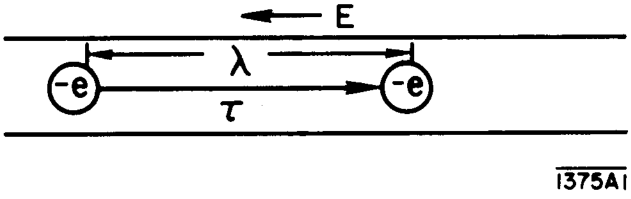

Fig. II. 1. Mean free path and time between collisions in a metal

When an electric field, $\mathscr{E}$, is applied to a metal, the electrons of charge, -e, are accelerated on the average for a time, τ, before being scattered. τ is called the relaxation time. The electrons are scattered by impurities, vacant sites, interstitial atoms, dislocation lines, grain boundaries, and other lattice defects. There is also scattering of the electrons by phonons. This can be visualized as density fluctuations due to thermal vibrations which, to the electrons, look like an increase or decrease of positive charge here and there. Phonon scattering is neglected in relating electrical and thermal conductivity as is done in this section. This is because when electrons are scattered through small angles by phonon interaction, the scattering is much more effective in producing a thermal than an electrical resistance.

Between collisions, the electrons gain an average drift velocity, v. After each scattering collision, the velocity, v, will be lost or so changed in direction as to be effectively 0 in the direction of the electric field.

$$v = \frac{-\nabla(\text{P.E.})\tau}{m} = \frac{-e\mathcal{E}\tau}{m} \tag{II.1}$$

where m is the effective mass of an electron in the metal. The electric current density, j, resulting from this transport of charge is:

$$j = n(-e)v = ne^2\mathcal{E}\tau/m \tag{II.2}$$

where n is the number density of free electrons.

∴ electrical conductivity,

$$\boxed{\sigma \equiv \frac{j}{\mathcal{E}} = ne^2\tau/m} \tag{II.3}$$

Thus one metal is a better conductor than another because it has a higher density of free electrons. Its electrical conductivity goes down with increasing high temperature because the time, τ, between collisions decreases.

B. Thermal Conductivity of Electrons

As we shall see, electrons make the dominant contribution to thermal conductivity of fairly pure metals in the normal state. If we think of the electrons in a metal as a free electron gas, then at a temperature, T, each electron has an average energy $\frac{3}{2}kT$ where k is the Boltzmann constant. At a temperature $T + \delta T$, the electron has an average energy $\frac{3}{2}k(T + \delta T)$. The excess energy, $\frac{3}{2}k\delta T$ causes the electron to diffuse into the colder region. If the metal has a thermal gradient, this will look somewhat like a potential gradient of magnitude $\nabla\left(\frac{3}{2}kT\right)$ acting like a "force" on each electron to move it from the region of high temperature to the region of lower temperature. This thermodynamic-like "force" gives rise to a drift of electrons down the metal controlled (for simplicity) as in the electrical case by collisions with lattice defects. The average drift velocity is

$$v = \frac{\nabla\left(\frac{3}{2}kT\right)\tau}{m} \quad . \tag{II.4}$$

Analogous to the electric current density, j, given by Eq. (II.2), the transport of thermal energy is equivalent to a heat current density,

$$\begin{aligned} q &= n\left(\frac{3}{2}kT\right)v \\ &= \left[\left(\frac{3}{2}\right)^2 nk^2 T\,\tau/m\right]\nabla T \; . \end{aligned} \tag{II.5}$$

The thermal conductivity is

$$\boxed{K \equiv \frac{q}{\nabla T} = \left(\frac{3}{2}\right)^2 nk^2 T\,\tau/m} \; . \tag{II.6}$$

Now, dividing Eq. (II.6) by Eq. (II.3):

$$\frac{K}{\sigma} = \left(\frac{3}{2}\right)^2 k^2 T/e^2 ,$$

and if we had been more careful and used statistical quantum mechanics, we'd have:

$$\boxed{\frac{K}{\sigma} = \frac{\pi^2}{3} k^2 T/e^2} . \qquad \text{(II. 7)}$$

Equation (II. 7) is the Wiedemann-Franz law. From it, we see why those metals that are good electrical conductors are also good thermal conductors, since the thermal conductivity is proportional to the electrical conductivity.

We must bear in mind that scattering by phonons was neglected in deriving the W-F law, so it will not be valid in the temperature range where phonon scattering is not a negligible process. We will see where this is shortly.

III. The Electronic Heat Capacity, Thermal Conductivity, and Diffusivity

Throughout this seminar, I shall use the term "heat capacity" to mean heat capacity at constant volume. Let us take a look at the density of occupied states of electrons in a metal as a function of energy, as shown in Fig. III. 1.

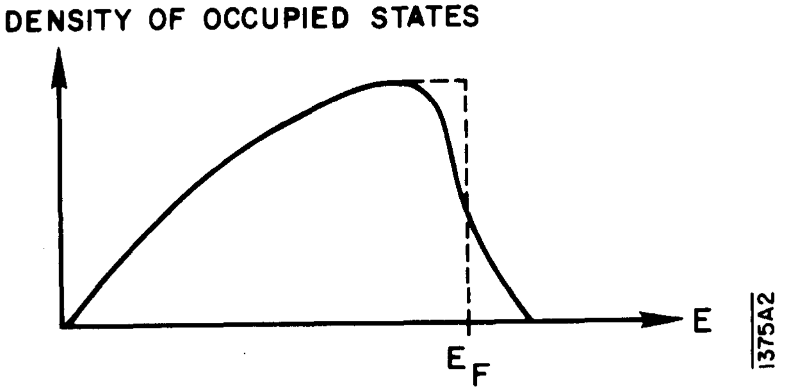

Fig. III. 1. Electron density of states as a function of energy

At 0° K, all the states are filled up to the Fermi energy, E_F. As the temperature, T, is raised, those electrons whose energies lie within a range kT from E_F will be excited to higher energy states. The electrons in lower states are essentially hemmed in by the filled states above them, as they are forbidden by the Pauli exclusion principle from sharing in these already filled higher states. Hence only those electrons that are within this energy range, kT from E_F, can be free in the classical sense. It is these electrons which contribute to the temperature dependence of the total energy, E, of the electrons, and hence which are responsible for the heat capacity, C'. To a good approximation, we can say that the fraction of the total number of electrons, N, that are excited is $\left(kT/E_F\right)$, and that each has acquired the extra classical energy $\frac{3}{2}kT$.

$$E = E_0 + \left(\frac{kT}{E_F}\right) N \left(\frac{3}{2}kT\right) = E_0 + \left(\frac{kT}{E_F}\right)(nV)\left(\frac{3}{2}kT\right) , \qquad \text{(III. 1)}$$

where E_0 is the total energy of the electrons at 0° K, and V is the volume of the metal.

$$C' = \frac{dE}{dT} = 3nVk^2T/E_F \qquad \text{(III.2)}$$

Had we been more careful, we'd have $\pi^2/2$ instead of 3 in Eq. (III.2). The heat capacity per unit volume,

$$C \equiv C'/V = \frac{\pi^2}{2} nk^2T/E_F \qquad \text{(III.3)}$$

Combining Eq. (III.3) algebraically with the W-F law (II.7), and substituting for σ from Eq. (II.3), we get:

$$K = \left(\frac{ne^2\tau}{m}\right)\left(\frac{1}{3e^2}\right)\left(\frac{2C}{n} E_F\right)$$

$$= \frac{1}{3} C\tau\left(\frac{E_F}{m/2}\right) \qquad \text{(III.4)}$$

$$\tau = \lambda/v \qquad \text{(III.5)}$$

where λ is the electron's mean free path between collisions.

$$\text{(III.5) in (III.4)} \Rightarrow K = \frac{1}{3}C\left(\frac{\lambda}{v}\right)\left(\frac{E_F}{m/2}\right) = \frac{1}{3}C\lambda v\left(\frac{E_F}{mv^2/2}\right) \qquad \text{(III.6)}$$

Now $\frac{1}{2}mv^2 \doteq E_F$ since E_F typically ~ 5 ev and the electrons do not increase their energy much above this.

$$\therefore \quad \boxed{K_e = \frac{1}{3} C_e\lambda_e v_e}, \qquad \text{(III.7)}$$

where the subscript e has been put in to denote explicitly that these are the electron's thermal conductivity, K_e; heat capacity per unit volume, C_e; mean free path, λ_e; and average velocity, v_e.

Equation (III. 7) can be derived more generally, and is valid over the whole temperature range. It is not limited by the Wiedemann-Franz law, nor by the particular temperature dependence of the heat capacity. We can see its generality by substituting (III. 7) into the thermal conduction equation for a rod of length d.

$$q = K\left(T_2 - T_1\right)/d \tag{III. 8}$$

$$= C\left(T_2 - T_1\right)\left(\frac{1}{3} v \frac{\lambda}{d}\right) \tag{III. 9}$$

Equation (III. 9) can be interpreted thus: $C\left(T_2 - T_1\right)$ is the excess energy density at one end of the rod with respect to the other end. This excess energy is propagated along the material with an effective transport velocity which is just 1/3 the carrier velocity reduced by the ratio of the mean free path to the length of the rod. Of course, the same ideas would pertain for other geometries, though not as clearly.

At very low temperatures, the product $\lambda_e v_e$ has essentially no temperature dependence. Since $C_e \propto T$ (III. 3), this implies $K_e \propto T$ (III. 7) at very low temperatures. We can also see this from the W-F law (II. 7), since the electrical resistivity, $\rho = 1/\sigma$, approaches a constant value ρ_0 at very low temperatures.

$$K_e = \left(\frac{\pi^2}{3} \frac{k^2}{e^2} \sigma_0\right) T = \left(\frac{\pi^2}{3} \frac{k^2}{e^2 \rho_0}\right) T \tag{III. 10}$$

The residual resistivity, ρ_0, is a result of impurities and other lattice defects in a metal. When the purity of two samples of a given metal is so high that it would be almost impossible to detect differences by any standard analytical technique, they can readily be compared by the electrical resistivity ratio. This is the ratio of the room temperature to the He temperature resistivity. While the residual resistivity, ρ_0, is very sensitive to the presence of impurity atoms, the room temperature resistivity is determined mainly by phonon (thermal vibrational) scattering for fairly pure metals.

The transient transport of heat (before the steady state thermal conductance equation can be applied) is described by

$$\frac{\partial T}{\partial t} = D\nabla^2 T \qquad \text{(III. 11)}$$

where D is the thermal diffusivity.

$$D = \frac{K}{C^*\rho} = \frac{\frac{1}{3}C\lambda v}{(C/\rho)\rho} = \frac{1}{3}\lambda v \qquad \text{(III. 12)}$$

where ρ is the mass density of the material, and the specific heat, $C^* = \frac{C'}{\rho\, v} = C/\rho$. Since λv is essentially temperature independent at very low temperatures, D is also.

IV. Relating the Phonon Specific Heat to the Phonon Thermal Conductivity

In the Superconducting Journal Club seminar I gave about a year ago, I derived the phonon (lattice) contribution to the heat capacity, C'_p, by Debye's method. From last year we know that at low temperature,

$$C'_p = AT^3 = \frac{dE}{dT} \qquad \text{(IV.1)}$$

$$\therefore \qquad E = \frac{1}{4}AT^4 = nV\langle E\rangle = nV(kT) \qquad \text{(IV.2)}$$

where n is the number density of phonons, V is the volume of the specimen, and $\langle E\rangle$ is the average energy per phonon.

$$\text{(IV.2)} \longrightarrow n = AT^3/4kV = C'_p/4kV = C_p/4k \qquad \text{(IV.3)}$$

where $C_p \equiv C'_p/V$ is the heat capacity per unit volume. Now the phonon heat current density is

$$q_p = n(k\lambda\nabla T)v = (C_p/4k)(k\lambda\nabla T)v = K_p\nabla T \qquad \text{(IV.4)}$$

where λ is the phonon mean free path, and K_p is the phonon thermal conductivity.

We can roughly define the phonon mean free path, λ, as the distance a phonon travels before its energy is reduced to 1/e of its original value. In a normal metal, phonons are scattered by other phonons, electrons, and lattice defects. In a superconductor (as in dielectrics), the scattering of phonons by electrons is negligible, particularly at very low temperatures. The phonon-phonon scattering arises due to the lattice anharmonicity as the phonons pass through regions of differing density. The lattice anharmonicity is increased when the lattice is strained, which increases the phonon scattering, and decreases the phonon thermal conductivity.

From Eq. (IV.4), we have

$$K_p = \frac{1}{4} C_p \lambda v ,$$

and if we had done it more carefully, we'd have

$$\boxed{K_p = \frac{1}{3} C_p \lambda_p v_p} \tag{IV.5}$$

where the subscript p has been put in to denote explicitly that these are the phonon quantities.

At very low temperatures, the product $\lambda_p v_p$ has negligible temperature dependence.

$$\therefore \quad K_p \propto T^3$$

for very low T.

V. Significance of the Heat Capacity and K_e/K_p

The general relationship $K = \frac{1}{3} C \lambda v$ has been variously credited to Drude and Debye. It should not be too surprising that the thermal conductivity is intimately related to the heat capacity. The heat capacity is quite a significant and basic quantity. That superconductors have an energy gap, and its magnitude, can be deduced from the superconducting specific heat. The relation between the critical magnetic field, H_c, and the critical temperature, T_c, $H_c \doteq H_0 \left[1 - \left(T/T_c\right)^2\right]$ can be derived from the difference between the superconducting and the normal heat capacities of the material. The fact that many different "soft" superconductors have approximately the same value of H_0/T_c results simply from the fact that they have approximately the same electronic specific heat in the normal state.

We are now in a position to estimate the relative contribution to the thermal conductivity of the electrons and the phonons in a fairly pure normal metal.

$$\frac{K_e}{K_p} = \frac{C_e v_e \lambda_e}{C_p v_p \lambda_p} = \frac{C_e v_e^2 \tau_e}{C_p v_p^2 \tau_p} \qquad (V.1)$$

where $\lambda = v\tau$.

At 300° K: C_p is 6 cal/mole - °K for most metals and has no temperature dependence as the temperature is increased; and C_e is typically $\sim 10^{-1}$ cal/mole- °K. $v_e \sim 10^8$ cm/sec, $\tau_e \sim 10^{-13}$ sec, $v_p \sim 3 \times 10^5$ cm/sec, and $\tau_p \sim 10^{-11}$ sec.

$$\therefore \quad \frac{K_e}{K_p} \sim 20 \text{ at } 300^\circ \text{K} . \qquad (V.2)$$

The theoretical prediction that in a pure metal at room temperature, the electron contribution to the thermal conductivity is about 20 times the phonon contribution is consistent with the experimental observation that at room temperature, metals have a thermal conductivity one or two orders of magnitude higher than those of solid crystalline dielectrics.

At low temperatures, $C_e = aT$ and $C_p = AT^3$, so

$$\frac{K_e}{K_p} = \frac{(aT)v_e\lambda_e}{(AT)^3 v_p \lambda_p} \quad . \tag{V.3}$$

To roughly see what this comes to, I have estimated some numbers for silver and copper at 2° K. For silver, $a \sim 0.6 \times 10^{-3}$ j/mole-°K^2, $A \sim 0.2 \times 10^{-3}$ j/mole-°K^4, $v_e \sim 1.4 \times 10^8$ cm/sec, $v_p \sim 3 \times 10^5$ cm/sec, $\lambda_e \sim 10^{-3}$ cm, and $\lambda_p \sim 10^{-1}$ cm.

$$\Rightarrow \frac{K_e}{K_p} \sim 4 \text{ for silver at } 2^\circ\text{ K} . \tag{V.4}$$

For copper, $a \sim 0.74 \times 10^{-3}$ j/mole-°K^2, $A \sim 0.5 \times 10^{-4}$ j/mole-°K^4, $v_e \sim 1.6 \times 10^8$ cm/sec, $v_p \sim 3 \times 10^5$ cm/sec, $\lambda_e \sim 10^{-3}$ cm, $\lambda_p \sim 10^{-1}$ cm.

$$\Rightarrow \frac{K_e}{K_p} \sim 20 \text{ for copper at } 2^\circ\text{ K.} \tag{V.5}$$

VI. Thermal Conductivity of Fairly Pure Normal Metals

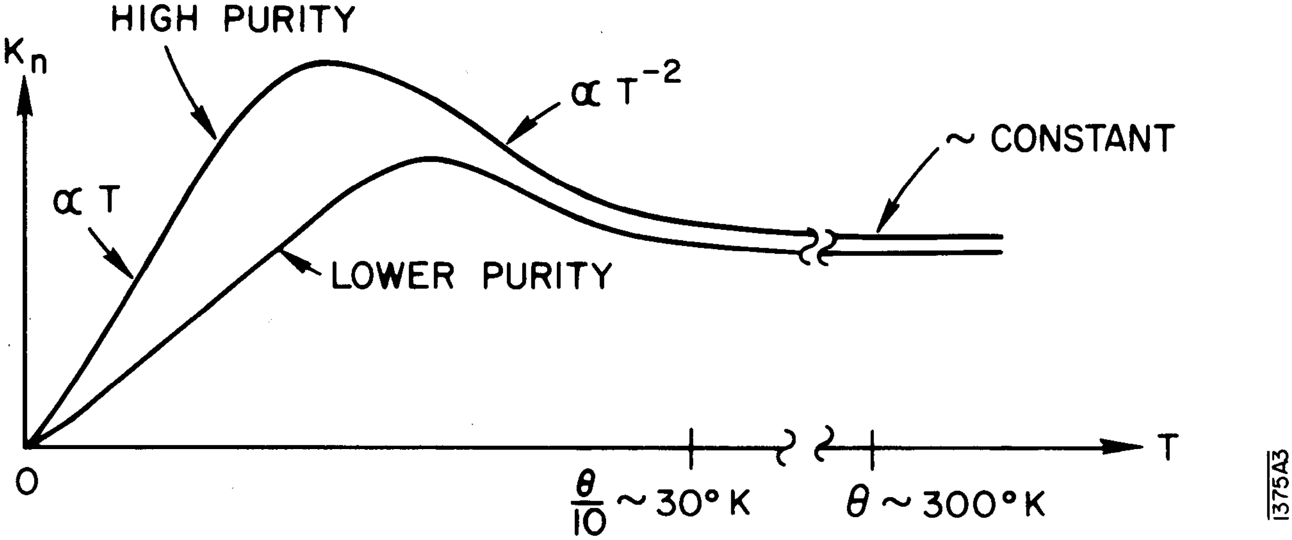

Fig. VI. 1. Thermal conductivity of two specimens of the same metal of different purity

Figure VI. 1 shows two typical curves for the thermal conductivity of metals as a function of temperature. The upper curve is for a metal of high purity and the lower curve is for the same metal of lower purity. The temperature dependence to the left of the maximum is linear. This can be seen from the W-F law or $K_e = \frac{1}{3} C_e \lambda_e v_e$. Since $\lambda_e v_e$ is essentially temperature independent and $C_e \propto T$, then $K_e \propto T$ in this region. The W-F law is valid here and similarly says that

$$K_e = \left(\frac{\pi^2}{3} \frac{k^2}{e^2}\right) \frac{T}{\rho_0} = LT/\rho_0 \qquad \text{(VI. 1)}$$

where ρ_0 is the residual electrical resistivity at very low temperature, and

$$L \equiv \left(\frac{\pi^2}{3} \frac{k^2}{e^2}\right) = 2.45 \times 10^{-8} \text{ watt-ohm/}^\circ\text{K}^2$$

is called the Lorenz number.

The W-F law is only valid where lattice defect scattering dominates, which is at very low temperatures where the phonon density and hence the phonon scattering of electrons is quite small and, as we shall see, at high temperatures. However, occasionally engineers take the W-F law to be valid over the whole temperature range and put in a variable Lorenz number to keep the relationship from breaking down. The danger in doing this is that the temperature dependence of L is variable, depending on the sample purity. The less pure the sample, the more defect scattering will dominate, and the less L will vary. Conversely, the more pure the sample, the more L will vary.

To the right of the thermal conductivity maximum, phonon scattering will dominate up to about the Debye temperature, θ, which is ~ 300° K for most metals. The W-F law is not valid in this region. As we saw in Section IV, the phonon density is $\propto T^3$. Therefore, the electron mean free path, $\lambda_e \propto T^{-3}$, since mean free paths are inversely proportional to the density of scatterers. The electron heat capacity per unit volume, $C_e \propto T$, and v_e is roughly constant. Therefore

$$K_e = \frac{1}{3} C_e \lambda_e v_e \Rightarrow K_e \propto T(T^{-3}) = T^{-2} . \qquad \text{(VI. 2)}$$

From the Debye temperature on, the average phonon energy cannot increase as the temperature is increased.

$$\text{Average Phonon Energy Maximum} = k\theta = h\nu_D \qquad \text{(VI. 3)}$$

where ν_D is the Debye frequency. This is the highest or cut-off frequency beyond which the lattice will not propagate a wave. This is essentially when the wavelength has decreased to the point that one-half wavelength is comparable to the lattice spacing. The total energy of the phonons increases as the temperature is raised because the number of phonons continues to increase. Since the average energy per phonon cannot increase, the phonons cannot take energy from the electrons beyond the Debye temperature. Hence the phonons are no longer effective in reducing the electron transport of thermal energy in the range $T > \theta$, and we are back to a lattice defect dominance. Thus the W-F law is valid in this region, and since $\rho \propto T$ here,

$$K_e = \frac{\pi^2}{3} \frac{k^2}{e^2} \frac{T}{\rho} \propto \frac{T}{T} = \text{constant} . \tag{VI.3}$$

Now that we have covered the whole temperature range, let's go back to the low temperature range and look at it more closely. In the range $0 < T < \theta/10$, we can express $1/K$ (called the thermal resistivity) as

$$\frac{1}{K} = \frac{\rho_0}{LT} + bT^2 . \tag{VI.4}$$

This can be derived theoretically by more refined arguments than those given here, or simply be considered as a best fit to the experimental data. If we solve for the maximum value of K, this occurs at a temperature,

$$T_m = \left(\frac{\rho_0}{2bL}\right)^{1/3} . \tag{VI.5}$$

Thus we see that for a given metal, the addition of impurities or other lattice defects increases the residual resistivity, ρ_0, and hence the temperature at which K is maximum, as well as reducing K. This explains the relative shapes of the two curves in Fig. VI.1.

To summarize: The conduction electrons and the lattice phonons contribute to thermal conductivity in a normal metal, and both contributions are limited by scattering from lattice imperfections, as well as by the mutual scattering of electrons and phonons. In a normal metal, at both high and low temperatures, the electrons make the dominant contribution to the thermal conductivity. At very low temperatures, the electron thermal conductivity is primarily limited by scattering from lattice point imperfections causing the conductivity to vary approximately linearly with temperature, and to decrease markedly with an increase in crystal defects.

VII. Thermal Conductivity of Superconductors and Dielectrics

In a superconductor, an increased density of "superconducting" electrons is found in the ground state as the temperature is decreased, with a corresponding decrease in the number of "normal" electrons. Since the electron energy gap is larger than the phonon energy at superconducting temperatures, the ground state electrons cannot interact to scatter (or be scattered by) the phonons, nor can they carry thermal energy. Perhaps the reason for this will be clearer after we look at the energy level diagram for a superconductor, as shown in Fig. VII. 1.

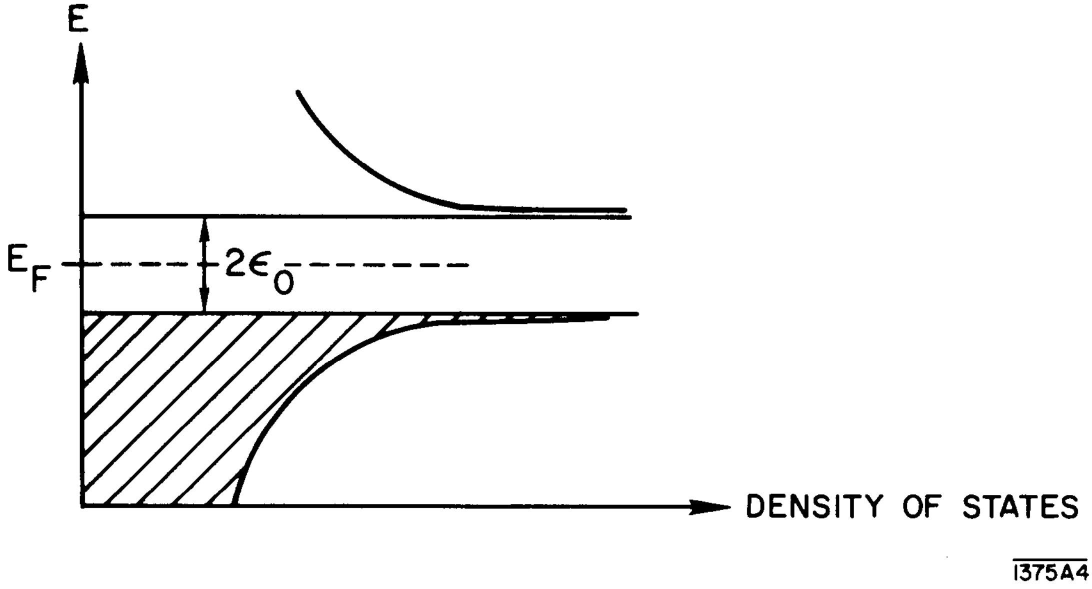

Fig. VII. 1. Electron density of states as a function of energy in a superconductor

The most singular aspect of the energy level diagram is the energy gap of width $2\,\epsilon_0$ centered about the Fermi energy, E_F. The gap represents a forbidden energy region, and results from quantum physics. It turns out that in the superconducting ground state, the electrons move around in pairs of opposite spin, and that it is not possible to break up a pair without introducing an energy of at least $2\epsilon_0$. Hence the configuration of electron pairs is a stable one at very low temperatures.

Until now, we have found that the thermal conductivity is higher, the higher the electrical conductivity. So it may come as a surprise to learn that a superconductor (which is as good an electrical conductor as one could hope for) is not a very good thermal conductor. The energy gap, $2\epsilon_0$, is typically $\sim 4kT_c$, where T_c is the critical temperature where the transition is made from the superconducting to the normal phase.

$$2\epsilon_0 \sim 4kT_c \sim 10^{-3}\ \mathrm{ev} \qquad \text{(VII. 1)}$$

At 1° K,

$$kT_1 \sim 10^{-4}\ \mathrm{ev} \qquad \text{(VII. 2)}$$

As long as $kT < 2\epsilon_0$, the superconducting electrons can't scatter or be scattered by phonons, nor can they carry thermal energy.

Let's see why this is so. At 0° K, all the energy states below the energy gap are filled. This is the shaded region in Fig. VII. 1. At temperature, $T < T_c$, a relatively small number of electrons near the energy gap are excited above it. (These act like normal electrons.) By the Pauli exclusion principle, the electrons whose energies lie below $\sim$ kT of the bottom of the energy gap cannot be excited into higher states as the higher states are already occupied. Nor can they go to lower states for the same reason. Those electrons that lie within $\sim$ kT of the bottom of the energy gap also have almost nowhere to go. They cannot lose energy because all the lower states are filled. There are relatively few empty states available between kT and the energy gap for them to go into. Those at the bottom of the energy gap cannot gain energy $\sim$ kT since this would put them in the forbidden region. They can only gain energy $\geq 2\epsilon_0$. Hence there can be no energy exchange between the superconducting electrons and the phonons, and only those electrons above the gap (which are now normal) can carry thermal energy. The number of these normal electrons decreases as T goes down from T_c.

Hence an increasing fraction of the thermal conduction is due to phonons as the temperature is lowered until they finally dominate the thermal conductivity in a superconductor. Not only is their relative contribution increased, but their absolute contribution is also increased due to the absence of electron scattering. Thus a given specimen has two sets of values of thermal conductivity with different functional dependence on temperature, depending on whether it is in the normal or superconducting state. In either case, the thermal conductivity is very sensitive to bulk defects, such as impurities, crystal boundaries, internal grain boundaries, dislocations, and strain, since all of these lead to phonon and "normal" electron scattering.

Since phonons provide the dominant thermal conductivity in dielectrics also, much of what will be said will apply to dielectrics as well as superconductors. (Many of the phonon processes we will discuss also occur in metals, but are overshadowed by the electron processes.) The main phonon scattering processes are:

1. Umklapp (phonon-phonon)
2. Boundary
3. Impurities and other point defects
4. Dislocation strain field
5. Normal electrons (for superconductors)

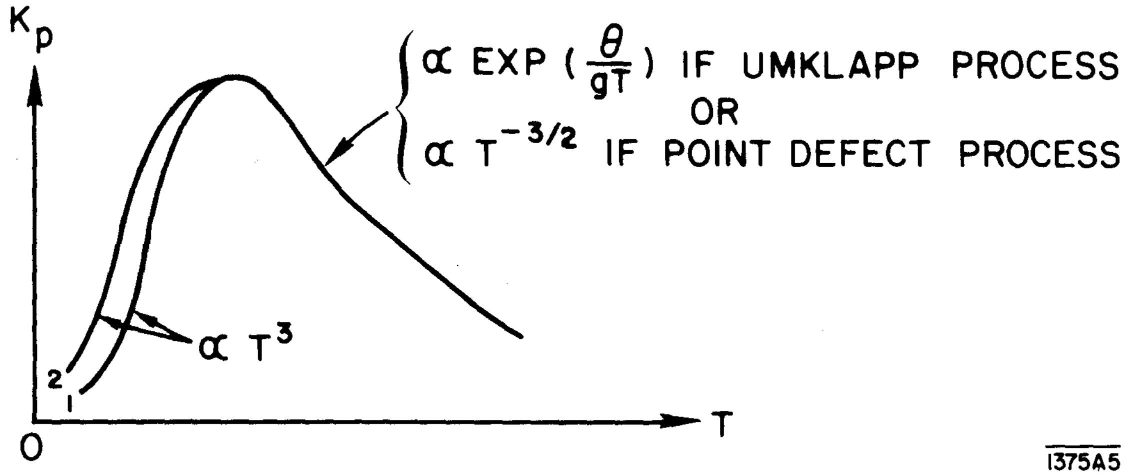

Fig. VII. 2. Thermal conductivity of crystalline dielectrics, or of superconductors at very low temperature

The umklapp process is a three-phonon process which occurs when two phonons of wave vectors k_1 and k_2 add together to yield a third phonon whose wave vector k_3 falls outside the zone boundary. This causes a reflection of k_3 much like the Bragg reflection of x-rays by the crystal lattice. This causes the resulting motion to be in approximately the opposite direction. This diminishes the energy flux in the original direction, giving rise to thermal resistance. "Umklapp" comes from the German, meaning "flipping over." The umklapp process is dominant in fairly pure, defect-free crystals to the right of the maximum in Fig. VII.2, where $K_p \propto \exp\left(\frac{\theta}{gT}\right)$. If the crystal has a significant number of impurities and other point defects, the exponential dependence will be suppressed and $K_p \propto T^{-3/2}$ in this region.

As the temperature is lowered, the mean free path of the phonons increases. As we go to temperatures to the left of the conductivity maximum, impurity atoms and other point defects do not much reduce K_p since they are now much smaller than the phonon wavelength and do little to scatter the phonons. At 300° K, the phonon mean free path, λ_p, is $\sim 10^{-5}$ to 10^{-6} cm in a metal, which is about the same as the electron mean free path, λ_e, in the metal. At low $T < \frac{\theta}{10}$, λ_e will increase to $\sim 10^{-3}$ cm, whereas λ_p will increase $\sim 10^{-1}$ cm or greater. At this point, $K_p \propto T^3$ from Eq. (IV.5), and will also be approximately $\propto$ smallest dimension of the specimen if it is a single crystal, or the size of the individual crystallites if it is polycrystalline. This is one of the reasons that a dielectric or superconducting monocrystal has a higher thermal conductivity than a polycrystal. This size effect is illustrated in Fig. VII.2, where the diameter of specimen 2 is greater than specimen 1, which are otherwise identical. Similarly, when boundary scattering becomes dominant over bulk scattering, the surface smoothness of the specimen is important if it is a monocrystal. If the surface is highly polished, K_p may be a factor of 2 to 3 greater at $\sim$ 2° K than if the surface had been rough-ground or etched. This is because the phonons will be specularly reflected rather than diffusely scattered at the surface. The behavior of a smooth and a rough surface is, respectively, like that of curves 2 and 1 in Fig. VII.2. One may think of the conductance of gas in pipes as an analogue to thermal conductance, with pressure as the analogue of temperature. At high pressure, the collision frequency in the gas is much higher than with the pipe walls. As the pressure is reduced and the mean free path increases, the collision frequency with the wall becomes much higher than between gas molecules, and the nature of the gas flow changes. Other scattering processes which further reduce K_p are the strain field induced either by external strain or in neighboring atoms near a dislocation; and by normal electrons.

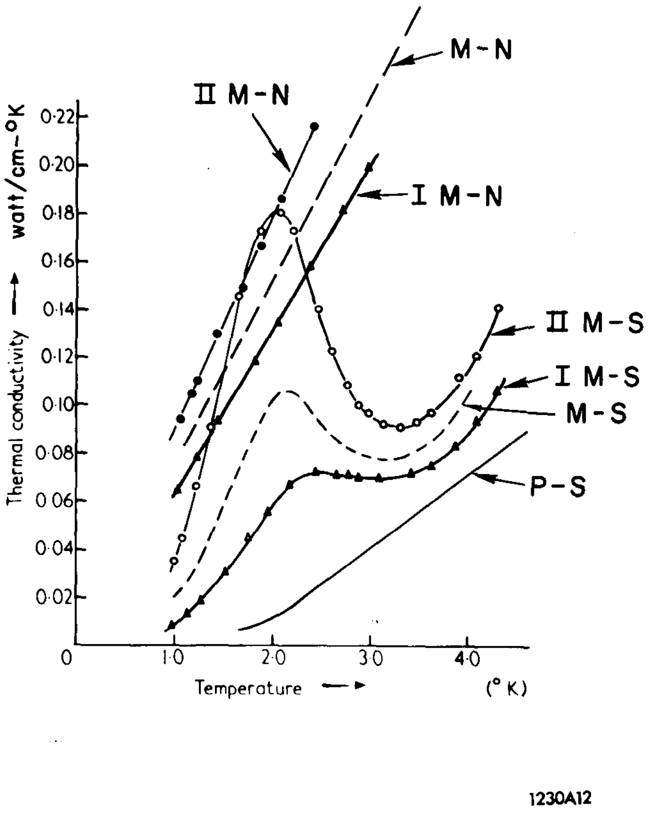

Fig. VII. 3. Thermal conductivity of normal and superconducting niobium under various conditions.

If we look at Fig. VII. 3, which is from Calverly et al.,[1] and has had a few curves added to it, we can see the thermal conductivity of niobium in the normal and superconducting states for various specimens and conditions. The curve

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[1] A. Calverly, K. Mendelssohn, and P. M. Rowell, Cryogenics 2, 26 (1961).

labeled NII is for a highly perfect monocrystal of niobium in the normal state. Curve SII is for this monocrystal in the superconducting state. The maximum comes at about 1.9° K, and is 0.18 watt/cm-°K. As Dick Neal has pointed out, operation on or to the right of the maximum, K_{max}, would be unstable because a slight temperature increase would lead to a decrease in K with a further increase in T. The curve labeled I is for a less perfect monocrystal which may also have some induced strain. The thermal conductivity is correspondingly reduced in both states. In the superconducting state, its peak is about 0.07 watt/cm-°K and is shifted to about 2.2° K. A superconducting polycrystalline sample is represented by the curve labeled P-S. At 1.9° K, its conductivity is down to 0.006 watt/cm-°K. I believe that the increase in K past the minimum is ascribable to the increase in the number of normal electrons as T increases.

Irradiation of a monocrystal much like specimen II, with a dose of 10^{18} fast neutrons/cm^2, reduces its thermal conductivity to the dashed curves labeled M-N and M-S. The original conductivity can be recovered in part by annealing the crystal at elevated temperatures. Single dislocations will anneal out (heal) readily. If dislocation clusters are formed, these will not heal as readily. The formation of isotopes will leave a permanent decrease in the thermal conductivity.

Care must be taken in measuring the normal state thermal conductivity, since the application of a magnetic field will increase the thermal resistivity just as it does the electrical resistivity. The relative increase is larger the lower the temperature. It is larger for transverse fields than for longitudinal, though a longitudinal field may at low values decrease the thermal resistivity. It is a larger effect for multivalent than for monovalent metals.

We could not expect to make the rf structure for the superconducting accelerator out of monocrystalline material. However, in addition to removing surface impurities and to some extent bulk impurities, the high temperature vacuum annealing may be expected to grow single crystals across the thickness of the wall with the grain boundaries appearing largely in the direction parallel to the heat flow across the cavity wall, rather than perpendicular to it. Though such a crystalline structure is not as favorable as a single monocrystal, it should have superior thermal conductivity to an ordinary polycrystal. The outlook for the disks in the cavities may not be as favorable if they are solid. However, if the cavities and hence the disks are hydroformed or if the disks are grooved on the outside so that the heat conduction is across the disk wall rather than from its iris to its outside diameter, then the outlook for the disks may also be favorable.

Therefore, in addition to making the usual thermal conductivity measurements on long thin rods, measurements should be made with both ordinary and recrystallized thin-walled cylinders across the wall thickness, though it may be a more difficult experiment. One possible engineering experiment which has occurred to me, and which I understand HEPL is also considering, is to measure the total temperature difference between the inside wall of a cavity and the liquid helium temperature as a function of the power input on the inside cavity wall. Due to Kapitza resistance (which we will consider shortly), this temperature difference is larger than that just across the cavity wall, so that the thermal conductivity of the wall itself would not be measured. However, this is just the engineering type of information we need.

One way of supplying the power would be to coat the inside cavity wall with a resistive coating and heat it electrically. Another way would be to support an electrical heater coaxially with the cavity along its length, and supply power to the inside cylindrical cavity wall by radiation. Either method has problems associated with it of putting in the power uniformly, of making sure that steady-state equilibrium is established and maintained, of measuring or inferring the inside wall temperature as a function of power dissipation, etc. I think the information to be gained is important enough that we should do this or some similar kind of simulation experiment.

VIII. Kapitza Resistance

Thermal boundary resistance is associated with heat flow across all solid-solid as well as all solid-liquid interfaces. There is a thermal boundary resistance between a superconductor and a normal metal which would be encountered for niobium or technetium deposited on copper. This has been measured between superconducting tin and copper.[2] Barnes and Dillinger found:

$$R_b = \frac{8.8}{T^3} \frac{\text{cm}^2\,{}^\circ\text{K}}{\text{watt}}$$

At 1.85° K:

$$R_b = \frac{8.8}{(1.85)^3} = \boxed{1.38 \frac{\text{cm}^2\,{}^\circ\text{K}}{\text{watt}}}$$

For peak flux:

$$\Delta T = R_K(\dot{Q}/A) = \left(1.38 \frac{\text{cm}^2\,{}^\circ\text{K}}{\text{watt}}\right)(16)(1.3 \times 10^{-3}\ \text{W/cm}^2) \doteq \boxed{.03^\circ\ \text{K}}$$

They also did it for superconducting lead on copper and found $R_b = (11.0/T^4)\text{cm}^2\text{-}{}^\circ\text{K/W}$. So if niobium or technetium on copper is similar to tin or lead on copper, this should be a negligible effect.

There is a temperature difference between a solid and the liquid helium next to it, due to the acoustic mismatch for the phonons traveling from the solid to the liquid. This is known as the Kapitza boundary effect, or Kapitza resistance, named after its discoverer (1941), P. L. Kapitza. This temperature difference is proportional to the heat flux from the solid to the liquid helium, and needs to be determined experimentally. Kapitza resistance is essentially a measure of the efficiency

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[2] L. J. Barnes and J. R. Dillinger, Phys. Rev. Letters <u>10</u>, 287 (1963).

with which the excess thermal energy of the solid is transmitted to the liquid. In metals, this energy primarily resides in electrons and these and the phonons must interact with the He across the interface to generate phonons in the liquid. This interaction depends on the acoustic impedance on both sides of the interface. In superconductors at very low temperatures, this energy resides primarily with the phonons. Therefore, one would expect a difference in the Kapitza resistance depending on whether the specimen is in the normal or superconducting state. One might expect a larger Kapitza resistance in the superconducting state, as there are then fewer coupling mechanisms, which is what is observed experimentally. The agreement between theory and experiment is still poor.[3] Theory predicts the Kapitza resistance to be one to two orders of magnitude higher than is measured.

The Kapitza resistance depends on the particular material as well as its surface condition, and is experimentally defined as $R_K = A \Delta T / \dot{Q}$, where A is the interfacial area, ΔT is the temperature difference, and $\dot{Q}$ is the heat flow. R_K is approximately $\propto T^{-3}$. The Kapitza resistance for the Nb-He II interface is not readily available. However, the upper limit Kapitza resistance for the Cu-He II interface of 10 cm^2- K/W at 1.85° K should be indicative. For an average heat flux of 1.3×10^{-3} $watt/cm^2$, this gives $\Delta T = 0.013°$ K, which is negligible. For an assumed duty cycle of 0.06, the peak flux is about 16 times this, giving $\Delta T = 0.21°$ K, which is not negligible. However, if there are spots where the field emission power dissipation is 0.1 $watt/cm^2$, the interface temperature difference may be as high as 1° K (assuming the worst case of a uniform heat flux through the wall), whereas the temperature difference across

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[3] G. L. Pollack, Rev. Mod. Phys. 41, 48 (1969).

a niobium wall of 0.25 cm thickness and 0.18 W/cm-°K thermal conductivity would be only 0.14° K. Large heat currents may lead to nonlinear and turbulence effects with a local destruction of the superfluidity of He II.

Despite the lack of experimental values for the Kapitza resistance of niobium, and the poor footing of the theory, an estimate can be made. We can relate the theory phenomenologically to the experimental values. The Khalatnikov theory[3] predicts that R_K is proportional to the atomic mass times the Debye temperature cubed of the particular solid. Using the range of 0.55 to 10 cm^2-°K/W for copper, this predicts a range of R_K for niobium of 0.246 to 4.48 cm^2-°K/W. However, using the range of 0.48 to 1.7 cm^2-°K/W for superconducting lead implies a range of 1.98 to 7.05 cm^2-°K/W for niobium.

IX. Acknowledgments

I wish to express my appreciation to Ed Garwin, Matt Allen, and Earl Hoyt for stimulating discussions.