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The Peierls condition and phonon-phonon scattering in metals

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A derivation is presented of the phonon-phonon scattering relaxation time for metals that takes explicit account of the Peierls condition, which states that normal phonon-phonon scattering cannot by itself equilibrate the phonon system. The expression for the phonon-phonon scattering relaxation time depends, in a complicated way, on the phonon-electron scattering relaxation time because of the coupling between phonon-electron scattering and phonon-phonon scattering that results from the Peierls condition. Analysis of the resulting expression shows that at low temperatures, the effect of the Peierls condition is to decrease phonon-phonon scattering very dramatically. The expression for the phonon-phonon scattering relaxation time is evaluated numerically for potassium as a function of temperature at a characteristic point in the Brillouin zone. It is found that at 1 K, the Peierls condition reduces phonon-phonon scattering for potassium by nearly an order of magnitude. A discussion is presented of the implication of these results for the phonon-drag contribution to the low-temperature electrical resistivity of the alkali metals. Comparison is made with other recent work.

I. INTRODUCTION

The phonon scattering relaxation time in solids has long been a subject of interest.[1,2] For pure insulators, phonon-impurity scattering is negligible and phonon-electron scattering is absent. Thus, the phonon system reaches thermal equilibrium by means of phonon-phonon scattering. By contrast, the phonons in pure metals are scattered both by other phonons and by electrons. The presence in metals of both phonon-phonon scattering and phonon-electron scattering leads to interesting results associated with the interplay between these two scattering mechanisms.

The calculation of the phonon scattering relaxation time for transport properties is complicated by the Peierls condition,[3] which states that normal phonon-phonon scattering cannot by itself equilibrate the phonon system. This does not imply that normal phonon-phonon scattering does not occur. In fact, at low temperatures, normal phonon-phonon scattering is much stronger than umklapp phonon-phonon scattering and therefore cannot simply be ignored. However, the correct calculation of the phonon-phonon scattering relaxation time must reduce to an infinite relaxation time in the absence of phonon scattering processes other than normal phonon-phonon scattering.

The Peierls condition on phonon-phonon scattering was first included in the calculation of a transport coefficient by Callaway,[4] who calculated the thermal conductivity of insulators. He incorporated the Peierls condition by means of the formulation given by Klemens.[5] According to Klemens,[5] the effect of normal phonon-phonon scattering is to relax each phonon state to a distribution function that is in thermal equilibrium except that the phonons are drifting with a given drift velocity. We here generalize the calculation of Callaway[4] to the case of metals, for which phonon-electron scattering is also present. It will be seen that the drift velocity depends strongly on the phonon-electron scattering time. We obtain an explicit expression for the phonon-phonon scattering relaxation time that takes full account of the Peierls condition and includes the important coupling to phonon-electron scattering. Evaluating the expression demonstrates that at low temperatures, the effect of including the Peierls condition is quite dramatic. Indeed, failure to take proper account of the Peierls condition leads to a serious overestimate of the importance of phonon-phonon scattering at low temperatures.

In Sec. II, the phonon-phonon scattering relaxation time is derived in terms of the drift velocity. In Sec. III, the drift velocity is calculated, displaying explicitly its dependence on the phonon-electron scattering relaxation time. The resulting expression for the phonon-phonon scattering relaxation time is analyzed qualitatively. Numerical results for the phonon-phonon scattering relaxation time for potassium are presented in Sec. IV. In Sec. V, the implications of these results are discussed for the phonon-drag contribution to the low-temperature electrical resistivity of the alkali metals. The summary follows in Sec. VI.

II. PHONON-PHONON SCATTERING RELAXATION TIME

The phenomenon of phonon-phonon scattering is generally analyzed within the relaxation-time approximation because of the practical impossibility of solving the full phonon Boltzmann equation. Within this framework, the temperature-dependent, phonon-phonon scattering relaxation time $\tau_{\text{ph-ph}}(\vec{q}\lambda)$ is defined by

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)_{\text{ph-ph scatt}} = -\frac{n(\vec{q}\lambda)-n^0(\vec{q}\lambda)}{\tau_{\text{ph-ph}}(\vec{q}\lambda)}\,, \tag{2.1}$$

where $n(\vec{q}\lambda)$ and $n^0(\vec{q}\lambda)$ are, respectively, the perturbed and thermal equilibrium phonon distribution functions for the phonon state of wave vector $\vec{q}$ and polarization λ.

Our goal[6] is to calculate $\tau_{\text{ph-ph}}(\vec{q}\lambda)$. It is necessary to treat separately the effects of normal phonon-phonon scattering and umklapp phonon-phonon scattering. The heart of the problem is the treatment of normal phonon-phonon scattering, for which one must insure that the Peierls condition is satisfied. Following Callaway,[4] the Peierls condition is incorporated into the analysis by requiring that in the presence of normal phonon-phonon scattering only, each phonon state will relax to a distribution function that is in equilibrium except for the fact that the phonons are drifting with a given velocity. Klemens[5] has pointed out that one may express this by writing

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)^{N}_{\text{ph-ph scatt}} = -\frac{n(\vec{q}\lambda)-n^0(\vec{q}\lambda;\vec{v})}{\tau_N(\vec{q}\lambda)}\,, \tag{2.2}$$

where

$$n^0(\vec{q}\lambda;\vec{v}) = \left(\exp\{[\hbar\omega_\lambda(\vec{q})-\vec{v}\cdot\vec{q}]/k_BT\}-1\right)^{-1}\,. \tag{2.3}$$

Here, $\vec{v}$ represents the drift velocity measured in units of $\hbar$. Although we have not explicitly included $\vec{q}\lambda$ indices for $\vec{v}$, the drift velocity in fact depends on both $\vec{q}$ and λ. However, the exact calculation of the $\vec{q}\lambda$ dependence of $\vec{v}$ for each phonon state requires an exact solution of the phonon Boltzmann equation, which we are determined to avoid. Therefore, some approximation must be made. This important point will be discussed in detail in the next section.

In a linear transport theory, the drift velocity is proportional to the magnitude of the external field acting on the phonons, be it a thermal gradient or an electric field. Therefore, we may expand the right-hand side of (2.3) and retain only the term linear in $\vec{v}$

$$n^0(\vec{q}\lambda;\vec{v}) = n^0(\vec{q}\lambda) + (\vec{q}\cdot\vec{v}) \times [n^0(\vec{q}\lambda)][n^0(\vec{q}\lambda)+1]/k_BT\,. \tag{2.4}$$

Equations (2.2)–(2.4) constitute the description of normal phonon-phonon scattering.

Umklapp phonon-phonon scattering is handled in the standard way, within the relaxation-time approximation

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)^{U}_{\text{ph-ph scatt}} = -\frac{n(\vec{q}\lambda)-n^0(\vec{q}\lambda)}{\tau_U(\vec{q}\lambda)}\,. \tag{2.5}$$

The relaxation times $\tau_N(\vec{q}\lambda)$ and $\tau_U(\vec{q}\lambda)$ can, in principle, be calculated for each phonon state (each $\vec{q}\lambda$) as a function of temperature. We again emphasize that one may not obtain $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ by merely adding $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ because such a simple addition would lead to the unphysical result that normal phonon-phonon scattering alone can equilibrate the phonon system.

The task before us is to determine $\tau_{\text{ph-ph}}(\vec{q}\lambda)$ as a function of $\tau_N(\vec{q}\lambda)$ and $\tau_U(\vec{q}\lambda)$. In order to calculate $\tau_{\text{ph-ph}}(\vec{q}\lambda)$, we turn to the phonon Boltzmann equation. Let us consider the specific conditions appropriate to the electrical resistivity, i.e., there is an electric field present but no thermal gradient. The electric field will cause both the electron distribution function $f(\vec{K})$ and the phonon distribution function $n(\vec{q}\lambda)$ to deviate from their thermal equilibrium values $f^0(\vec{K})$ and $n^0(\vec{q}\lambda)$, respectively,

$$\begin{aligned} f(\vec{K}) &= f^0(\vec{K}) - \phi_{\text{el}}(\vec{K})\,\frac{\partial f^0(\vec{K})}{\partial E(\vec{K})}\,,\\ n(\vec{q}\lambda) &= n^0(\vec{q}\lambda) - \phi_{\text{ph}}(\vec{q}\lambda)\,\frac{\partial n^0(\vec{q}\lambda)}{\partial \hbar\omega_\lambda(\vec{q})}\,, \end{aligned} \tag{2.6}$$

where $E(\vec{K})$ is the energy of the electron in state $\vec{K}$ and $\omega_\lambda(\vec{q})$ is the angular frequency of the $\vec{q}\lambda$ phonon. The electron and phonon functions, $\phi_{\text{el}}(\vec{K})$ and $\phi_{\text{ph}}(\vec{q}\lambda)$, represent the deviations from thermal equilibrium that are caused by the electric field. The functions $\phi_{\text{el}}(\vec{K})$ and $\phi_{\text{ph}}(\vec{q}\lambda)$ are determined from the coupled electron and phonon Boltzmann equations. For the conditions under consideration, electric field present but no thermal gradient, the relaxation-time-approximation phonon Boltzmann equation becomes

$$-\int\!\!\int d^3K_1\, d^3K_2\, [\phi_{\text{el}}(\vec{K}_1)+\phi_{\text{ph}}(\vec{q}\lambda)-\phi_{\text{el}}(\vec{K}_2)]P_{\text{ph-el}}(\vec{K}_1,\vec{q}\lambda;\vec{K}_2) + \left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)_{\text{ph-ph scatt}} = 0\,. \tag{2.7}$$

The second term in (2.7) is the total contribution due to phonon-phonon scattering and is given either by (2.1) or by the sum of (2.2) and (2.5). The first term of (2.7) is the contribution due to phonon-electron scattering. We are considering a pure metal for which phonon-impurity scattering is unimportant. The quantity $P_{\text{ph-el}}(\vec{K}_1,\vec{q}\lambda;\vec{K}_2)$ is the probability that the $\vec{q}\lambda$-phonon will cause an electron to be scattered from the initial electron state $\vec{K}_1$ to the final electron state $\vec{K}_2$. The explicit expression for $P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)$ for the case of a spherical Fermi surface is well known.[1,2]

Equation (2.7) can be simplified by introducing the phonon-electron scattering relaxation time.[7] The first term of (2.7), which represents the total change in $n(\vec{q}\lambda)$ due to phonon-electron scattering, can be written as the sum of two terms.

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)_{\text{ph-el scatt}} = -\phi_{\text{ph}}(\vec{q}\lambda)\int\int d^3K_1\, d^3K_2\, P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2) + \int\int d^3K_1\, d^3K_2\, [\phi_{\text{el}}(\vec{K}_2) - \phi_{\text{el}}(\vec{K}_1)] P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)\ . \tag{2.8}$$

In the first term of the right-hand side of (2.8), the electron system is in equilibrium $[\phi_{\text{el}}(\vec{K}) = 0]$ and the phonon system is not in equilibrium $[\phi_{\text{ph}}(\vec{q}\lambda) \neq 0]$, whereas in the second term, the phonon system is in equilibrium and the electron system is not in equilibrium. To define a phonon-electron scattering relaxation time, one imagines the case where the electron system is in equilibrium. This leads to the following equation for the phonon-electron scattering relaxation time $\tau_{\text{ph-el}}(\vec{q}\lambda)$:

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)^{\text{ph-el scatt}}_{\phi_{\text{ph}}(\vec{q}\lambda) \neq 0;\ \phi_{\text{el}}(\vec{K}) = 0} = -\frac{n(\vec{q}\lambda) - n^0(\vec{q}\lambda)}{\tau_{\text{ph-el}}(\vec{q}\lambda)} = -\phi_{\text{ph}}(\vec{q}\lambda)\int\int d^3K_1\, d^3K_2\, P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)\ . \tag{2.9}$$

By means of the second equation of (2.6), one obtains

$$\frac{1}{\tau_{\text{ph-el}}(\vec{q}\lambda)} = \frac{k_B T}{n^0(\vec{q}\lambda)[n^0(\vec{q}\lambda) + 1]} \times \int\int d^3K_1\, d^3K_2\, P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)\ . \tag{2.10}$$

We now show that $\tau_{\text{ph-el}}(\vec{q}\lambda)$ is independent of temperature by examining the temperature dependence of the integral in (2.10). The energy integration[2] of the δ function in energy contained in $P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)$ introduces a factor $[n^0(\vec{q}\lambda) + 1]/k_B T$. Moreover, $P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)$ contains[2] an explicit factor of $n^0(\vec{q}\lambda)$. These factors exactly cancel the coefficient multiplying the integral in (2.10). To emphasize this point, it is convenient to introduce a "normalized" phonon-electron scattering probability

$$C_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2) = P_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2) k_B T / \times [n^0(\vec{q}\lambda)][n^0(\vec{q}\lambda) + 1]\ , \tag{2.11}$$

where the "normalized" probability $C_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)$ is essentially the phonon-electron scattering probability per phonon. Inserting (2.11) into (2.10) yields

$$\frac{1}{\tau_{\text{ph-el}}(\vec{q}\lambda)} = \int\int d^3K_1\, d^3K_2\, C_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)\ , \tag{2.12}$$

where the integral, and hence $\tau_{\text{ph-el}}(\vec{q}\lambda)$, is independent of temperature. Combining (2.12) and (2.11) with (2.7) leads to

$$\left(-\frac{\phi_{\text{ph}}(\vec{q}\lambda)}{\tau_{\text{ph-el}}(\vec{q}\lambda)} + D(\vec{q}\lambda)\right)\frac{n^0(q\lambda)[n^0(q\lambda) + 1]}{k_B T} + \left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)_{\text{ph-ph scatt}} = 0\ , \tag{2.13}$$

where we have introduced the notation

$$D(\vec{q}\lambda) \equiv \int\int d^3K_1\, d^3K_2 [\phi_{\text{el}}(\vec{K}_2) - \phi_{\text{el}}(\vec{K}_1)] \times C_{\text{ph-el}}(\vec{K}_1, \vec{q}\lambda; \vec{K}_2)\ . \tag{2.14}$$

It is clear that $D(\vec{q}\lambda)$ is independent of temperature by the same analysis that led to the conclusion that $\tau_{\text{ph-el}}(\vec{q}\lambda)$ is independent of temperature.

Having cast the phonon Boltzmann equation into the convenient form (2.13), we proceed to the calculation of $\tau_{\text{ph-ph}}(\vec{q}\lambda)$. Inserting (2.1) into (2.13) and combining with the second equation of (2.6) yields the expression for the phonon function

$$\phi_{\text{ph}}(\vec{q}\lambda) = D(\vec{q}\lambda)/[\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda) + \tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)]\ . \tag{2.15}$$

Equivalently, we may insert (2.2) and (2.5) into (2.13) to obtain an alternative expression for the phonon function

$$\phi_{\text{ph}}(\vec{q}\lambda) = \tau_{\text{com}}(\vec{q}\lambda) D(\vec{q}\lambda) + (\vec{v}\cdot\vec{q})\tau_N^{-1}(\vec{q}\lambda)\tau_{\text{com}}(\vec{q}\lambda)\ , \tag{2.16}$$

where we have introduced the "combined" phonon scattering relaxation time

$$\tau^{-1}_{\text{com}}(\vec{q}\lambda) = \tau_N^{-1}(\vec{q}\lambda) + \tau_U^{-1}(\vec{q}\lambda) + \tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)\ . \tag{2.17}$$

Equating these two equivalent expressions for $\phi_{\text{ph}}(\vec{q}\lambda)$, (2.15) and (2.16), yields the desired result

$$\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)=\tau^{-1}_{\text{com}}(\vec{q}\lambda)[1+(\vec{v}\cdot\vec{q})\tau_N^{-1}(\vec{q}\lambda)D^{-1}(\vec{q}\lambda)]^{-1}$$
$$-\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)\ . \qquad (2.18)$$

Equation (2.18) does not complete the calculation of $\tau_{\text{ph-ph}}(\vec{q}\lambda)$ because there remains to determine the drift velocity $\vec{v}$ by application of the Peierls condition. This is the subject of Sec. III.

III. DRIFT VELOCITY

Even without a detailed calculation of the drift velocity, one may already draw important conclusions from Eq. (2.18). First, consider the assumption of zero drift velocity. Such an assumption corresponds to ignoring entirely the Peierls condition. This can be seen from the fact that if one assumes (incorrectly) that normal phonon-phonon scattering could equilibrate the phonon system, then Eq. (2.2) should describe the relaxation of the phonon system to true thermal equilibrium. In other words, in Eq. (2.2) we should replace $n^0(\vec{q}\lambda;\vec{v})$ by $n^0(\vec{q}\lambda)$, which is equivalent to setting $\vec{v}=0$. Assuming that $\vec{v}=0$ and combining (2.18) and (2.17) yield immediately that $\tau^{-1}_{\text{ph-ph}}(\vec{q})$ is just the sum of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$.

To examine the effect of taking into account the fact that the drift velocity is actually not zero, it is convenient to rewrite Eq. (2.18) into the form

$$\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)=\tau_U^{-1}(\vec{q}\lambda)+\tau_N^{-1}(\vec{q}\lambda)$$
$$\times\left(1-\frac{(\vec{v}\cdot\vec{q})D^{-1}(\vec{q}\lambda)\tau^{-1}_{\text{com}}(\vec{q}\lambda)}{1+(\vec{v}\cdot\vec{q})D^{-1}(\vec{q}\lambda)\tau_N^{-1}(\vec{q}\lambda)}\right). \qquad (3.1)$$

Since $\vec{v}\cdot\vec{q}$ will be shown to be positive, one sees immediately from (3.1) that the effect of a nonzero drift velocity is to reduce $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ by multiplying $\tau_N^{-1}(\vec{q}\lambda)$ by a factor smaller than unity. This is exactly what is expected on physical grounds. The Peierls condition reduces the effectiveness of normal phonon-phonon scattering as a mechanism for equilibrating the phonons, thus leading to an over-all reduction in $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. However, the magnitude of the reduction in $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ is very difficult to estimate from (3.1). When scattering mechanisms are present other than normal phonon-phonon scattering (in the present case, these include umklapp phonon-phonon scattering and phonon-electron scattering), then $\tau_{\text{ph-ph}}(\vec{q}\lambda)$ depends on $\tau_N(\vec{q}\lambda)$ and on $\tau_U(\vec{q}\lambda)$ in a quite complicated way. It is this dependence that we now calculate.

The drift velocity is determined by explicit application of the Peierls condition. The requirement that there be no net change in the total crystal momentum due to normal phonon-phonon scattering is expressed[4] as follows:

$$\sum_\lambda\int d^3q\,\vec{q}\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)^N_{\text{ph-ph scatt}}=0\ , \qquad (3.2)$$

where the integral is taken over the entire Brillouin zone. The dependence of $[\partial n(\vec{q}\lambda)/\partial t]^N_{\text{ph-ph scatt}}$ on $\vec{v}$ is given by Eqs. (2.2)–(2.4).

Strictly speaking, the drift velocity depends on $\vec{q}$ and λ. But, since $\vec{v}$ is to be determined from the single condition (3.2), one must in some way average over this $\vec{q}\lambda$ dependence. In other words, one is to view $\vec{v}$ as a weighted average over $\vec{q}$ and λ of the true $\vec{q}\lambda$-dependent drift velocity $\vec{v}(\vec{q}\lambda)$. However, it is important how the averaging process is carried out. For example, one may certainly not just treat $\vec{v}(\vec{q}\lambda)$ as a constant in evaluating (3.2).

The optimal procedure for performing the averaging of $\vec{v}(\vec{q}\lambda)$ can be seen by studying the simpler case of the lattice thermal conductivity of insulators. Callaway[4] has shown that $\vec{q}\cdot\vec{v}$ must be proportional to the driving field, in that case, the thermal gradient. In the present case, in the absence of a thermal gradient, the phonon system is driven from thermal equilibrium by an effective field. The phonon function $\phi_{\text{ph}}(\vec{q}\lambda)$ must of course be proportional to this effective driving field, since it is the effective driving field that causes the phonon system to deviate from thermal equilibrium. Examination of Eq. (2.15) for $\phi_{\text{ph}}(\vec{q}\lambda)$ leads one to identify $D(\vec{q}\lambda)$ with the effective field that drives the phonon system from thermal equilibrium. Thus, according to Eq. (2.16), we must have

$$\vec{q}\cdot\vec{v}(\vec{q}\lambda)\propto D(\vec{q}\lambda)\ . \qquad (3.3)$$

The result is not unexpected if one recalls that $D(\vec{q}\lambda)$ depends linearly on $\phi_{\text{el}}(\vec{K})$ from Eq. (2.14) and notes that $\phi_{\text{el}}(\vec{K})$ is proportional to the applied electric field. Thus, (3.3) implies that $\vec{v}(\vec{q}\lambda)$ is proportional to the electric field. This is required on physical grounds because it is the electric field that drives the electron system from thermal equilibrium and the nonequilibrium electron system in turn drives the phonon system from thermal equilibrium via the electron-phonon interaction.

Having established the proportionality between $\vec{q}\cdot\vec{v}(\vec{q}\lambda)$ and $D(\vec{q}\lambda)$, we return to the problem of averaging $\vec{v}(\vec{q}\lambda)$. The simplest approximation is to assume a $\vec{q}\lambda$-independent constant of proportionality in (3.3). Thus, we write

$$\vec{q}\cdot\vec{v}(\vec{q}\lambda)=\alpha D(\vec{q}\lambda)\ , \qquad (3.4)$$

where our approximation consists of assuming that the true function $\alpha(\vec{q}\lambda)$ depends sufficiently weakly on $\vec{q}\lambda$ that it may be replaced by a constant. The constant α is to be determined from the Peierls condition, as given by (3.2).

To evaluate (3.2), and thus determine α, we rewrite the expression for normal phonon-phonon scattering, given by Eq. (2.2), by means of Eqs. (2.4), (2.6), (2.17), and (3.4). A short calculation yields

$$\left(\frac{\partial n(\vec{q}\lambda)}{\partial t}\right)^{N}_{\text{ph-ph scatt}} = \frac{D(\vec{q}\lambda)n^0(\vec{q}\lambda)[n^0(\vec{q}\lambda)+1]}{\tau^{-1}_{\text{com}}(\vec{q}\lambda)k_BT} \times\{-1+\alpha[\tau_U^{-1}(\vec{q}\lambda)+\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)]\}. \tag{3.5}$$

Multiplying (3.2) by the applied electric field $\vec{F}$ and inserting (3.5) leads to the explicit expression for α,

$$\frac{1}{\alpha} = \left(\sum_\lambda \int d^3q\,(\vec{q}\cdot\vec{F})D(\vec{q}\lambda)n^0(\vec{q}\lambda)[n^0(\vec{q}\lambda)+1]\tau_{\text{com}}(\vec{q}\lambda)\tau_N^{-1}(\vec{q}\lambda)[\tau_U^{-1}(\vec{q}\lambda)+\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)]\right) \times\left(\sum_\lambda \int d^3q\,(\vec{q}\cdot\vec{F})D(\vec{q}\lambda)n^0(\vec{q}\lambda)[n^0(\vec{q}\lambda)+1]\tau_{\text{com}}(\vec{q}\lambda)\tau_N^{-1}(\vec{q}\lambda)\right)^{-1}. \tag{3.6}$$

Note that α is independent of the magnitude of $\vec{F}$. From the fact that $D(\vec{q}\lambda)\propto\vec{F}$, it follows that for metals having cubic symmetry, α is also independent of the direction of $\vec{F}$.

Equations (2.18), (3.4), and (3.6) provide an explicit expression for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ as a function of temperature and hence constitute the central result of this paper. Before turning to the numerical calculation of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for a particular metal, it is instructive to examine the general expression in some detail. To this end, it is convenient to define certain weighted averages. We introduce the weight factor $R(\vec{q}\lambda)$, given by

$$R(\vec{q}\lambda)\equiv(\vec{q}\cdot\vec{F})D(\vec{q}\lambda)n^0(\vec{q}\lambda)\times[n^0(\vec{q}\lambda)+1]\tau_{\text{com}}(\vec{q}\lambda)\tau_N^{-1}(\vec{q}\lambda), \tag{3.7}$$

and denote normalized weighted averages over $\vec{q}$ and λ as follows:

$$\langle A\rangle\equiv\left(\sum_\lambda\int d^3q\,A(\vec{q}\lambda)R(\vec{q}\lambda)\right)\times\left(\sum_\lambda\int d^3q\,R(\vec{q}\lambda)\right)^{-1}. \tag{3.8}$$

In terms of such averages, one sees from (3.6) that

$$\alpha^{-1}=\langle\tau_U^{-1}\rangle+\langle\tau^{-1}_{\text{ph-el}}\rangle\,. \tag{3.9}$$

Inserting (3.4) and (3.9) into (3.1) yields, after a short calculation,

$$\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)=\tau_U^{-1}(\vec{q}\lambda)+\tau_N^{-1}(\vec{q}\lambda)\times\left(1-\frac{\tau_N^{-1}(\vec{q}\lambda)+\tau_U^{-1}(\vec{q}\lambda)+\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)}{\tau_N^{-1}(\vec{q}\lambda)+\langle\tau_U^{-1}\rangle+\langle\tau^{-1}_{\text{ph-el}}\rangle}\right). \tag{3.10}$$

Equation (3.10) is particularly useful for general discussion. We first note that the factor in the large parentheses multiplying $\tau_N^{-1}(\vec{q}\lambda)$ results from the Peierls condition. This factor makes clear the strong interplay between the various scattering processes. Both phonon-electron scattering and umklapp phonon-phonon scattering are seen to play an important role in determining by how much the contribution of normal phonon-phonon scattering is reduced by the Peierls condition. Also, it is immediately clear from (3.10) that if only normal phonon-phonon scattering is present, then $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ vanishes.

One should note the particular importance of the $\vec{q}\lambda$ dependence of $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$. If these phonon scattering processes were independent of $\vec{q}\lambda$, then $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)=\langle\tau^{-1}_{\text{ph-el}}\rangle$ and $\tau_U^{-1}(\vec{q}\lambda)=\langle\tau_U^{-1}\rangle$. Thus, the second term of (3.10) would vanish identically, regardless of the relative strengths of the different phonon scattering processes. It is the $\vec{q}\lambda$ dependence of the various phonon scattering processes that leads to the complex interdependence of $\tau_N^{-1}(\vec{q}\lambda)$, $\tau_U^{-1}(\vec{q}\lambda)$, and $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$ in determining $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$.

There is an important special circumstance which merits discussion. Consider the case for which normal phonon-phonon scattering is much stronger than the other phonon scattering processes. For this case, the second term in the large parentheses of (3.10) is nearly unity, and therefore, the contribution of $\tau_N^{-1}(\vec{q}\lambda)$ to $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ is greatly reduced. This is precisely what happens for metals at low temperatures. For certain ranges of $\vec{q}$ and for a specific polarization, $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$ can be very small. Moreover, $\tau_U^{-1}(\vec{q}\lambda)$ decays exponentially[1-3] with temperature at low temperatures, leaving $\tau_N^{-1}(\vec{q}\lambda)$ as the dominant of the three scattering processes. However, because of the Peierls condition, the resulting $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ is much smaller than $\tau_N^{-1}(\vec{q}\lambda)$. Under these circumstances, the effect of the Peierls condition is particularly dramatic.

IV. NUMERICAL CALCULATIONS

In Sec. III, a detailed qualitative discussion was given of the effect of the Peierls condition on the values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. We now complete the analysis by presenting the results of a quantitative calculation of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for a particular metal. For our illustrative metal we choose potassium, both for its intrinsic interest as the characteristic alkali metal, as well as for the more prosaic reason that the required input data for the calculation are available for potassium.

Values for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ as a function of temperature are obtained from Eqs. (2.18), (3.4), and (3.6). As input data, one requires the values[8] of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$. Also, one needs to know the electron function $\phi_{\text{el}}(\vec{K})$ in order to calculate $D(\vec{q}\lambda)$, as given by Eq. (2.14). First, consider the function $\phi_{\text{el}}(\vec{K})$. For the low-temperature regime, say up to about 5 K for potassium, impurity scattering is the dominant scattering process for the electrons even for the purest available samples of potassium. This implies[2] that $\phi_{\text{el}}(\vec{K})=\vec{K}\cdot\hat{F}$, where $\hat{F}$ is a unit vector in the direction of the applied electric field. This form for $\phi_{\text{el}}(\vec{K})$ is exact for low temperatures. However, explicit calculations for the alkali metals[9-11] show that using this form for $\phi_{\text{el}}(\vec{K})$ for higher temperatures introduces an error of only a few percent which decreases rapidly with increasing temperature. Therefore, for our illustrative purposes, it is certainly adequate to use $\phi_{\text{el}}(\vec{K})=\vec{K}\cdot\hat{F}$ for all temperatures.

The other required input data are the values for $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$. Albers, Bohlin, Roy, and Wilkins[8] (ABRW) have recently carried out a first-principle calculation of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ for potassium as a function of $\vec{q}$ and λ for several different temperatures. Unfortunately, the accuracy of the ABRW values of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ is limited. ABRW[8] have called attention to the difficulties inherent in a first-principle calculation and they conclude that their values of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ should be viewed as a first estimate only. This limited accuracy presents no problem for our purposes, since we are primarily interested in illustrating the effect of the Peierls condition, rather than in obtaining definitive values for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. Thus, we may view the ABRW values for $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ as exact for a hypothetical metal and we shall calculate the corresponding values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$.

ABRW[8] present results for $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ in detail (for all three polarizations over a range of temperatures) only for the [110] direction, whereas, according to Eq. (3.6), the calculation of the drift velocity requires values for all directions of $\vec{q}$. We overcome this difficulty by relying on the finding of ABRW[8] that there is only a weak dependence of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ on the direction of $\vec{q}$. This suggests that one may neglect this weak dependence altogether. We have tested this approximation by explicit calculation and find that a factor-of-2 variation in $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ with direction of $\vec{q}$ only changes the values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ by a few percent. This result is not unexpected in view of the fact that, according to (3.6), the angular integrations over $\vec{q}$ of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ appear in both numerator and denominator and therefore tend to cancel. Alternatively, one may approach the matter by reverting to our hypothetical metal, which is now imagined to have angular-independent values of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$.

To summarize, we take the values of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ directly from ABRW.[8] We note that the data are presented in terms of half widths $\Gamma_\lambda(\vec{q})$, which are defined by $2\Gamma_\lambda(\vec{q})=\tau^{-1}(\vec{q}\lambda)$. Moreover, the quantity $\Gamma_\lambda^{\text{tot}}(\vec{q})$ of ABRW is defined as sum of $\Gamma_\lambda^N(\vec{q})$ and $\Gamma_\lambda^U(\vec{q})$. The values are assumed independent of the direction of $\vec{q}$ and the values for intermediate $|\vec{q}|$ are obtained by quadratic interpolation. This approximation is certainly consistent with the accuracy of the calculated values of $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$.

In Table I, we present our calculated values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for all three polarizations λ for a specific value of $\vec{q}$ for three temperatures (1, 9, and 30 K) for which values for $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ are given. We choose $\vec{q}=0.2(2\pi/a)(1,1,0)$ as a characteristic point in the Brillouin zone. We have verified that other choices of $\vec{q}$ lead to quite similar results. The values for $\tau_N^{-1}(\vec{q}\lambda)$ and $\tau_U^{-1}(\vec{q}\lambda)$ are taken from Table III of Ref. 8.

The last column of our Table I relates the calculated values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$, which include the

TABLE I. Temperature dependence of the normal, umklapp, and phonon-phonon scattering relaxation times for potassium for the point $\vec{q}=0.2(2\pi/a)(1,1,0)$. The last column gives the ratio of the phonon-phonon scattering relaxation time that includes the Peierls condition to that obtained upon ignoring the Peierls condition.

Polarization (λ)	T (K)	τ_U^{-1}	τ_N^{-1}	$\tau^{-1}_{\text{ph-ph}}$	$\dfrac{\tau^{-1}_{\text{ph-ph}}}{\tau_N^{-1}+\tau_U^{-1}}$
		(10^9 rad/sec)			
Longitudinal	9	0.8	7	3	0.4
(L)	30	2	26	14	0.5
Higher transverse (T_1)	1	0.06	2	0.1	0.05
	9	0.1	4	2	0.5
	30	4	22	16	0.6
Lower transverse (T_2)	1	0.04	0.4	0.09	0.2
	9	1	9	3	0.3
	30	12	64	30	0.4

Peierls condition, with the values which would be obtained if one were to ignore the Peierls condition. Recall that without the restriction of the Peierls condition, $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ would be given by just the sum of $\tau^{-1}_{N}(\vec{q}\lambda)$ and $\tau^{-1}_{U}(\vec{q}\lambda)$. Thus, the last column of the table gives a quantitative measure of the effect of the Peierls condition expressed as the ratio of the true $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ to that obtained by ignoring the Peierls condition. One sees that the effect of the Peierls condition is quite dramatic at very low temperatures, leading to nearly an order-of-magnitude decrease in $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ at 1 K. Therefore, we may conclude that it is imperative to include the Peierls condition in the calculation of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. Failure to do so at low temperatures will lead to values for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ that are very much larger than the true values.

V. LOW-TEMPERATURE ELECTRICAL RESISTIVITY

The role played by $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ in the calculation of the electrical resistivity of the alkali metals was discussed by Bailyn[12] many years ago. The central result of Bailyn is an exact expression for the phonon-drag contribution to the electrical resistivity $\rho(T)$. We may write

$$\rho(T)=\rho_{\text{equil}}(T)-\rho_g(T)\,, \tag{5.1}$$

where $\rho_{\text{equil}}(T)$ is the resistivity for the case where the phonon system is in thermal equilibrium and $\rho_g(T)$ gives the reduction in the resistivity due to phonon drag. Recast into our notation, the Bailyn expression[12] for $\rho_g(T)$ is

$$\rho_g(T)=A\sum_{\lambda}\int\int\int d^3K_1\,d^3K_2\,d^3q\,[\phi_{\text{ph}}(\vec{q}\lambda)] \times[\phi_{\text{el}}(\vec{K}_2)-\phi_{\text{el}}(\vec{K}_1)]P_{\text{el-ph}}(\vec{K}_1,\vec{q}\lambda;\vec{K}_2)\,, \tag{5.2}$$

where A is a known constant. The term $\rho_g(T)$ depends on phonon scattering through its dependence on the phonon function $\phi_{\text{ph}}(\vec{q}\lambda)$, which in turn depends on $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$, as given by Eq. (2.15) which we repeat here for convenience

$$\phi_{\text{ph}}(\vec{q}\lambda)=D(\vec{q}\lambda)/[\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)+\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)]\,. \tag{2.15}$$

Recall that $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$ is independent of temperature, whereas $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ is strongly temperature dependent. At very low temperatures, $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda) \ll \tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$, implying that $\phi_{\text{ph}}(\vec{q}\lambda)$, and hence $\rho_g(T)$, is large and independent of phonon-phonon scattering. This may be called the "drag limit." At high temperatures, phonon-phonon scattering is so strong that $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)\to\infty$, implying that $\phi_{\text{ph}}(\vec{q}\lambda)\to 0$. In this limit, according to (5.2), $\rho_g(T)$ becomes negligible and one obtains the usual high-temperature result that $\rho(T)$ does not contain any contribution due to phonon drag.

The important question is: Below what temperature does one enter the drag limit in which one may ignore phonon-phonon scattering in the calculation of $\rho(T)$? ABRW recently addressed themselves[8,13] to this question, basing their answer on their calculated values of $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. However, ABRW calculated $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ without including the Peierls condition and thus obtained unphysically large values for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. Therefore, one cannot accept the ABRW conclusion[8,13] that, for potassium "the drag limit may not be reached until temperatures at least as low as 1 K."

One may estimate roughly the temperature marking the onset of the drag limit from the following considerations. It has been shown[11] that the phonons which dominate the low-temperature resistivity lie on the lower transverse (T_2) branch in the vicinity of the [110] direction. For low q, the T_2 phonons that lie *exactly* in the [110] direction do not, of course, contribute to $\rho(T)$ at all because of the polarization-vector factor,[11] but nevertheless, because of their low frequency, the T_2 phonons in the vicinity of the [110] direction dominate $\rho(T)$. The precise dominant direction is a function of temperature; the lower the temperature, the closer the dominant direction is to the [110] direction. Therefore, it is sufficient for the low-temperature calculation of $\rho(T)$ to calculate $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for the T_2 phonon branch for directions near the [110] direction.

We obtain the values of $\tau^{-1}_{N}(\vec{q}\lambda)$ and $\tau^{-1}_{U}(\vec{q}\lambda)$ for intermediate temperatures, between 1 and 9 K, by interpolating the ABRW results, which are quoted in our Table I. The accuracy of the interpolation can be verified at $T\simeq 4$ K from the curve given by APRW[8] in their Fig. 9(c). One may use the results that refer to the [110] direction, since $\tau^{-1}_{N}(\vec{q}\lambda)$ and $\tau^{-1}_{U}(\vec{q}\lambda)$ depend only weakly[8] on the direction of $\vec{q}$. This by no means implies that $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ is nearly isotropic. According to Eq. (3.10), $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ depends on $\tau^{-1}_{\text{ph-el}}(\vec{q}\lambda)$, which has a very marked dependence on the direction of $\vec{q}$. It follows, therefore, that $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ also depends significantly on direction. Thus, we must calculate $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for directions near, but not parallel to, the [110] direction. For each temperature, we calculate $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ for the T_2 phonons for the direction that dominates $\rho(T)$. Carrying out this calculation leads to the value of about 5 K for the temperature marking the onset of the drag limit for potassium. We emphasize that this temperature is only a rough estimate because of the limited accuracy of the ABRW values for $\tau^{-1}_{N}(\vec{q}\lambda)$ and $\tau^{-1}_{U}(\vec{q}\lambda)$, to say nothing of the interpolation we have made between 1 and 9 K.

This resulting temperature of about 5 K, may be compared with experiment in the following way. One calculates $\rho(T)$ without $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$, call it $\rho^0_{\text{calc}}(T)$, and compares with the $\rho_{\text{expt}}(T)$ data. For very low temperatures, well within the drag limit, $\rho^0_{\text{calc}}(T)$ is in excellent agreement[14] with $\rho_{\text{expt}}(T)$. As the temperature increases, one begins to observe deviations between $\rho^0_{\text{calc}}(T)$ and $\rho_{\text{expt}}(T)$, which are well accounted for by inserting into the calculation an appropriate expression for $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$. Carrying out such an analysis in detail[15] leads to a temperature of 6 K for the onset of the drag limit, in close agreement (fortuitously close, in fact) with the rough estimate of about 5 K given above. Thus, for $T<6$ K, one may safely ignore $\tau^{-1}_{\text{ph-ph}}(\vec{q}\lambda)$ in the calculation of $\rho(T)$ for potassium. Above 6 K, one must include phonon-phonon scattering in the calculated phonon-drag contribution to the electrical resistivity.

VI. SUMMARY

In this paper, we derive an expression for the phonon-phonon scattering relaxation time for metals that takes explicit account of the Peierls condition, which states that normal phonon-phonon scattering cannot by itself equilibrate the phonon system. Analysis of the resulting expression shows that the Peierls condition reduces the effect of the normal phonon-phonon scattering relaxation time by a factor that depends on certain weighted averages of the umklapp phonon-phonon scattering relaxation time and the phonon-electron scattering relaxation time. At very low temperatures, normal phonon-phonon scattering is very much reduced, leading to a dramatic reduction in the phonon-phonon scattering relaxation time. A quantitative calculation was carried out for potassium. The numerical values obtained for the phonon-phonon scattering relaxation time confirm the results of the qualitative discussion.

The implications of these results are discussed for the low-temperature electrical resistivity of the alkali metals. It is shown that the Peierls condition increases by about a factor of five the temperature below which one need not include phonon-phonon scattering in the calculation of the phonon-drag contribution to the electrical resistivity. In particular, these results justify the neglect of phonon-phonon scattering in resistivity calculations for potassium below 6 K.

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