(1970).

[14] N. C. Halder, J. Chem. Phys. 52, 5450 (1970).

[15] P. Jena, T. P. Das, G. D. Gaspari, and N. C. Halder, Phys. Rev. B 3, 2158 (1971).

[16] C. P. Slichter, *Principles of Magnetic Resonance* (Harper & Row, New York, 1963).

[17] E. Clementi, IBM J. Res. Develop. 9, 2 (1965).

[18] A. O. E. Animalu and V. Heine, Phil. Mag. 12, 1249 (1965).

[19] N. W. Ashcroft and J. Lekner, Phys. Rev. 145, 83 (1966); J. E. Enderby and D. M. North, J. Phys. Chem. Liquids 1, 1 (1968).

[20] P. Jena and N. C. Halder, Phys. Rev. B 4, 711 (1971).

[21] A. L. Ritter and J. A. Gardner, Phys. Rev. B 3, 46 (1971).

[22] S. D. Silverstein, Phys. Rev. 130, 912 (1963).

PHYSICAL REVIEW B VOLUME 4, NUMBER 8 15 OCTOBER 1971

Electron-Phonon Umklapp Scattering Processes in the Low-Temperature Ultrasonic Attenuation and Electrical Resistivity of Potassium*

P. N. Trofimenkoff† and J. W. Ekin‡

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14850

(Received 27 May 1971)

The theoretical treatment by Rice and Sham of the electron-phonon contribution to the attenuation of ultrasound in the local limit and the ideal (phonon-limited) electrical resistivity of potassium is extended to include a careful evaluation of the role of umklapp scattering processes at low temperatures. The results have been explicitly separated into normal and umklapp scattering components and the rapid decrease of the umklapp components, and hence the total, at very low temperatures, is emphasized for both the attenuation and resistivity. The results obtained from a realistic phonon model are compared in one case with those from an isotropic Debye-like model with a quadratic dispersion relationship and purely longitudinal and transverse polarization vectors.

I. INTRODUCTION

Several recent developments in the study of the ultrasonic attenuation due to electron-phonon interactions and the ideal electrical resistivity at low temperature have suggested a more thorough examination of the role played by electron-phonon normal and umklapp scattering processes in potassium.

In a recent paper, Rice and Sham[1] calculated the amplitude attenuation constant by the pseudopotential method for a longitudinal ultrasonic wave in potassium in the local limit where the phonon-limited electron mean free path is short compared with the impressed ultrasound wavelength. They compared their calculations (corrected to account for sample dimensions as opposed to the infinite medium, and for the polycrystalline nature of the samples) to the experimental data taken by Natale and Rudnick[2] on polycrystalline potassium samples at three ultrasonic frequencies in the temperature region $T \approx 2-20$ °K. Rice and Sham found that the theoretical values underestimated the measured attenuation constant by a factor approaching 2.

In recent years, measurements[3] of the electrical resistivity of potassium at very low temperatures have indicated that there are deviations from the T^5 temperature dependence predicted by the Bloch-Grüneisen formula which neglects electron-phonon umklapp scattering processes. Similar effects have been observed in sodium by Woods[4]; the interpretation in sodium, however, is somewhat obscured by the martensitic phase transformation. Very recently a quantitative comparison[5] of accurate resistivity measurements with accurate theoretical calculations (whose details are presented in this paper) has shown that the complicated temperature dependence in potassium at low temperatures can be completely accounted for in terms of a "freezing out" of umklapp processes.

Peierls[6] was the first to point out that for a metal in which the Fermi surface does not touch the Brillouin-zone boundary there exists a minimum phonon wave vector required for umklapp processes to be possible. At a temperature below the energy corresponding to this minimum wave vector, the umklapp component of the resistivity will decrease nearly exponentially owing to the effect of the Bose-Einstein distribution function for the thermal phonons. The relative importance of normal and umklapp processes has been estimated in the past.[7] In particular, Bailyn pointed out that owing to the anisotropy in the sound velocities the effect suggested by Peierls would occur at rather low temperatures in potassium.

In this paper a thorough analysis of the relative importance of electron-phonon normal and umklapp scattering processes in determining the low-temperature behavior of both the ultrasonic attenuation and the ideal electrical resistivity of potassium is presented. With various model pseudopotentials and a realistic phonon spectrum the contributions due to umklapp processes are accurately evaluated down to temperatures less than 1 °K. This work goes beyond that of Hasegawa,[7] whose tabulation of the normal and umklapp separation for the electrical resistivity is given only above 8 °K; Rice and Sham[1] present only the total components for the attenuation and resistivity.

II. THEORY

The early experiments of Bömmel[8] on the ultrasonic attenuation in single crystals of lead and those of MacKinnon and Bömmel[9] on polycrystalline and single-crystal samples of tin spurred several theoretical analyses[10] of the attenuation in normal metals in addition to the earlier work of Akhiezer.[11] The essential idea involved is that the local electron distribution is not in thermal equilibrium. The distribution is relaxed via electron-ion collisions. In addition an internal electric field is established. For longitudinal waves the field is due to the out-of-phase motion of the electrons and ions; for shear waves the field is an inductive one. A wave propagating through the metal is damped owing to the combined effect of the collisions and the field. A wave moving through an infinite medium in the z direction takes the form

$$A = A_0\, e^{-\alpha z}\, e^{i(Kz-\omega t)} \tag{1}$$

with phase velocity $c=\omega/K$, where ω is the angular frequency, K is the wave number, t is the time, and α is the amplitude attenuation constant.

In this paper only the local limit will be considered. For longitudinal waves in the local limit the attenuation constant α_L is

$$\alpha_L = \tfrac{4}{15}\, n\, E_F\, \omega^2 \tau / \xi\, c_L^3 \;, \tag{2}$$

where n is the valence-electron density, ξ is the density of the metal, E_F is the Fermi energy ($E_F = \hbar^2 k_F{}^2/2m$, where $\hbar$ is Planck's constant divided by 2π, k_F is the Fermi wave number, and m is the electron mass),[12] c_L is the phase velocity of the wave, and τ is a relaxation time for the electrons. The corresponding expression for the attenuation constant α_T for shear waves is

$$\alpha_T = \tfrac{1}{5}\, n\, E_F\, \omega^2 \tau / \xi\, c_T^3 \;, \tag{3}$$

where c_T is the phase velocity of the shear wave. In early comparisons with experiment, τ was identified with the relaxation time appropriate for the electrical resistivity τ_1. It was pointed out by Bhatia and Moore[13] that in the local limit ($Kl_1 \ll 1$, $l_1 = v_F \tau_1$, where v_F is the Fermi velocity) the relaxation time τ appropriate in Eqs. (2) and (3) is not the same as that for the electrical resistivity, but is an effective relaxation time τ_2.

Potassium occurs in a body-centered cubic structure with a valence of one electron per atom. The Fermi surface is believed to be very nearly spherical and not to touch the Brillouin-zone boundary as is shown, for example, by studies of the de Haas-van Alphen effect.[14] Thus, a one orthogonalized-plane-wave treatment should be sufficient to describe the electron-phonon interaction. In the approximation of a local pseudopotential $W(\vec{q})$ and a spherical Fermi surface, the relaxation times τ_l ($l=1, 2$) are given by[1,13,15]

$$\frac{1}{\tau_l} = \frac{\Omega_0 N(0)}{4k_F^2}\;\frac{\beta\hbar}{M}\sum_\lambda \int_{<2k_F} d^3q \times \frac{|\vec{\epsilon}(\vec{q};\lambda)\cdot\vec{q}\,|^2\,|W(\vec{q})|^2}{q\,(1-e^{-\beta\hbar\omega(\vec{q};\lambda)})(e^{\beta\hbar\omega(\vec{q};\lambda)}-1)}\,[1-P_l(\cos\theta)] \;, \tag{4}$$

where Ω_0 is the volume per ion, $N(0)$ is the single-spin electron density of states per unit volume at the Fermi surface, $N(0) = mk_F/2\pi^2\hbar^2$, M is the ion mass, and $\beta = 1/k_B T$, where k_B is Boltzmann's constant. The vector $\hbar\vec{q}$ is the momentum transferred to an electron scattered from the initial state $\vec{k}_i$ to the final state $\vec{k}_f$ (both on the Fermi surface), $\vec{q} = \vec{k}_f - \vec{k}_i$. The P_l are the Legendre polynomials and $\cos\theta = \vec{k}_i \cdot \vec{k}_f / k_F^2$. The phonons are described in a repeated zone scheme; they are specified by their frequencies $\omega(\vec{q};\lambda)$ and polarization vectors $\vec{\epsilon}(\vec{q};\lambda)$ for wave vector $\vec{q}$ and polarization branch λ (one longitudinal, two transverse). The integral extends over a sphere of radius $2k_F$.

The vector $\vec{q}$ in the phonon variables may be read as $\vec{q}$ reduced to the first Brillouin zone by a reciprocal-lattice vector. That part of the integral for which $\vec{q}$ lies in the first Brillouin zone with no reduction is classified as the normal contribution; for $\vec{q}$ values outside the first Brillouin zone the contribution is the umklapp component. For the realistic phonon model the phonon frequencies and eigenvectors were generated from a five-nearest-neighbor force-constant model as fitted to the inelastic neutron scattering and sound velocity data of Cowley, Woods, and Dolling.[16] The numerical work was lessened by evaluating the frequencies and eigenvectors in the irreducible $\frac{1}{48}$ of the first Brillouin zone[17] and transforming these values outward in a cone subtended by the basic $\frac{1}{48}$ to a radius $2k_F$. The lattice parameter a for the conventional cubic cell was taken to be $a = 5.225$ Å throughout.[18]

As a test of the effect of the phonon model, the relaxation times were also calculated with a Debye-like model. A quadratic phonon-dispersion relationship was used with one purely longitudinal and two degenerate purely transverse branches. The phonon parameters for this model are those given for potassium by Allen and Cohen.[19]

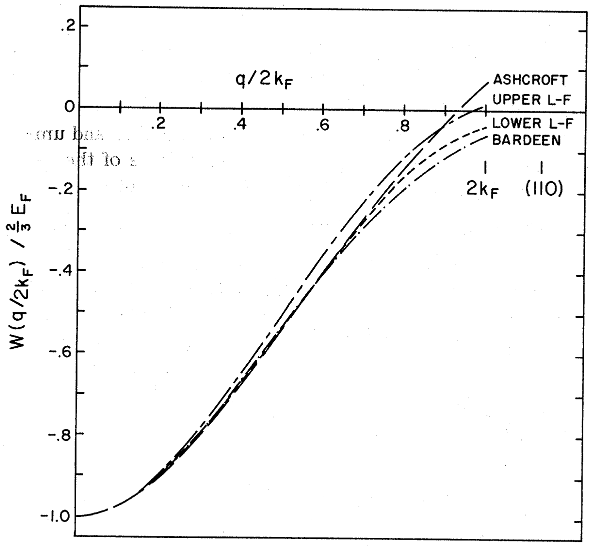

FIG. 1. Pseudopotentials of Bardeen (Ref. 20), Ashcroft (Ref. 21), and upper and lower pseudopotentials of Lee and Falicov (Ref. 22).

III. RESULTS AND DISCUSSION

In Fig. 1 the Bardeen,[20] Ashcroft,[21] and two other pseudopotentials relating to the work of Lee and Falicov[22] are shown for comparison. The essential points to note are the low-wave-number behavior and the behavior near $2k_F$. At low wave numbers all the pseudopotentials are nearly degenerate; this is the region that determines the normal component and hence the low-temperature behavior of the relaxation times. The region near $2k_F$ is the important region for the umklapp processes.

In Fig. 2 the results for (a) the ultrasonic-attenuation and (b) the resistivity-relaxation times are presented for the case of the well-known and easily reproduced Ashcroft pseudopotential; the purpose of Fig. 2 is to explicitly depict the manner of addition of the normal and umklapp components to produce the total for both the resistivity and attenuation. In order to graphically illustrate the role of umklapp processes in the attenuation relative to the resistivity an effective (for purposes of comparison only) resistivity calculated using τ_2 is defined:

$$\rho_l = m/ne^2\tau_l \tag{5}$$

for $l = 1,\ 2$, where e is the electronic charge. The results are presented as coefficients of the T^5 dependence. As can readily be seen, the total ultrasonic attenuation is affected far less by umklapp processes than is the total electrical resistivity. In the ultrasonic attenuation the umklapp component lies well below the normal component and the total is only slightly enhanced over the normal component. The total resistivity, however, is strongly influenced by the umklapp component with the umklapp component becoming dominant above ≈ 2.5 °K. In fact, although the normal component of the attenuation exceeds that of the electrical resistivity, the umklapp component of the attenuation is smaller than that of the electrical resistivity. As will be seen shortly, these considerations apply with only a few modifications to the other pseudopotentials.

The relatively weak influence of the umklapp component in the attenuation is explained[1] by the angular weighting factor $1 - P_l(\cos\theta)$ of Eq. (4). For the resistivity it is $1 - P_1(\cos\theta) = 1 - \cos\theta = |\vec{q}\,|^2/2k_F{}^2$; for the attenuation $1 - P_2(\cos\theta) = \frac{3}{2} \times (1 + \cos\theta)\ (1 - \cos\theta) = 3(1 - |\vec{q}\,|^2/4k_F{}^2)(|\vec{q}\,|^2/2k_F{}^2)$. The additional factor is $3(1 - |\vec{q}\,|^2/4k_F{}^2)$ for the attenuation; this factor is small at $|\vec{q}\,| \lesssim 2k_F$ which is

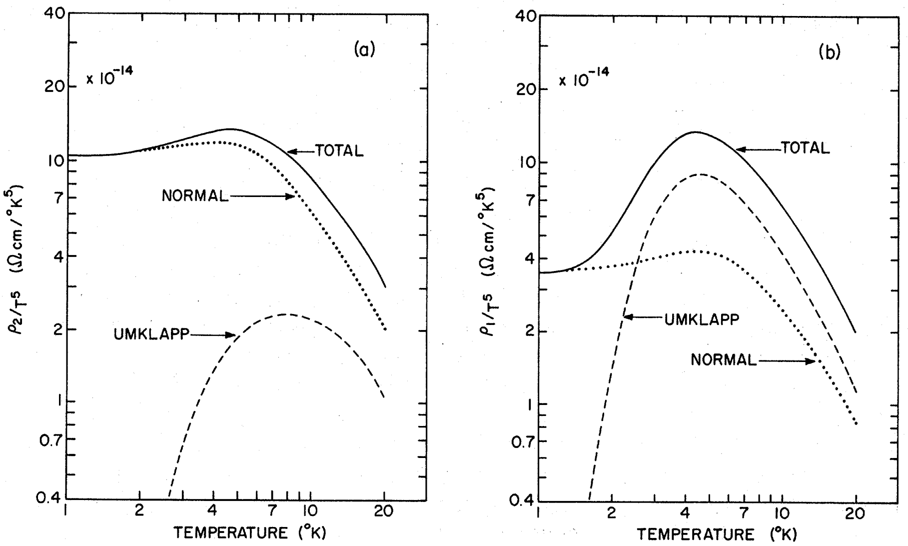

FIG. 2. Relaxation times plotted as the coefficient of the T^5 power of resistivities for (a) ultrasonic attenuation and (b) electrical resistivity for the Ashcroft pseudopotential. The normal, umklapp, and total components are given in each case.

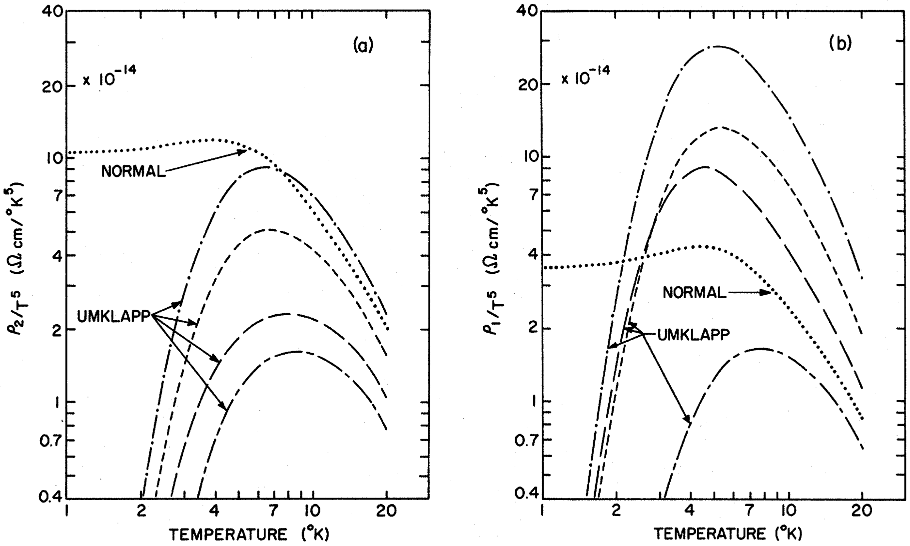

FIG. 3. Normal and umklapp components of the relaxation times plotted as coefficients of the T^5 power of resistivities for (a) ultrasonic attenuation and (b) electrical resistivity. In both (a) and (b) the umklapp components refer to the Bardeen, upper and lower Lee-Falicov, and Ashcroft pseudopotentials (represented as in Fig. 1).

the important region for umklapp processes.

It is interesting to note that the normal component itself shows a decrease steeper than T^5 in the low-temperature region $\approx 4-2\,^\circ$K. This dependence is due, to the dispersion in the phonon energy-versus-wave-number relationship.

In Fig. 3 the normal and umklapp components for (a) the ultrasonic attenuation and (b) the resistivity of all four pseudopotentials are depicted. As previously mentioned, the normal component depends on the small-wave-number-part of the pseudopotential and is virtually identical for all four forms. The umklapp components are directly related to the behavior of the pseudopotentials at $|\vec{q}| \lesssim 2k_F$ [compare the sizes of the umklapp components to the absolute value of the pseudopotentials at $|\vec{q}| = 2k_F$ remembering that it is the square of the pseudopotential that determines the resistivity through Eqs. (4) and (5)]. Thus it is the sensitivity of the umklapp component to the form of the pseudopotential that is responsible for the difference between the low-temperature resistivity values calculated with different pseudopotentials; the same statement holds for the attenuation.

In Fig. 4 the total coefficients of the T^5 dependence are shown for (a) the attenuation and (b) the resistivity. The solid curve in Fig. 4(b) represents the experimental data of Ref. 5. The agreement between theory and experiment both in the temperature dependence and the position of the maximum is excellent. It is reemphasized that the different low-temperature dependences for the different pseudopotentials result entirely from the difference in the umklapp components. If theory is to be correct, then the ultrasonic attenuation from experiment should show a behavior similar to that of the resistivity in the range 5–2.5 °K. The effect calculated in the attenuation, however, is visibly less pronounced.

To point out the importance of an accurate phonon

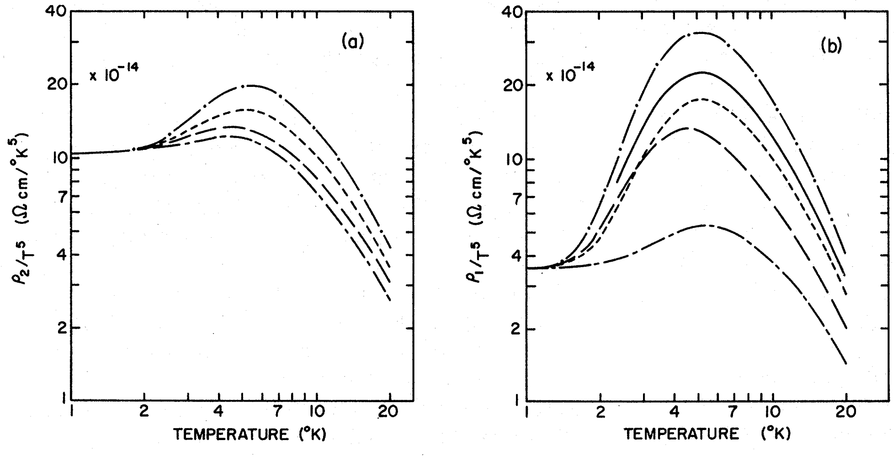

FIG. 4. Total relaxation times plotted as coefficients of the T^5 power of resistivities for (a) ultrasonic attenuation and (b) electrical resistivity. In both (a) and (b) the reference is to the Bardeen, upper and lower Lee-Falicov, and Ashcroft pseudopotentials (represented as in Fig. 1). The solid curve in (b) represents the data of Ref. 5.

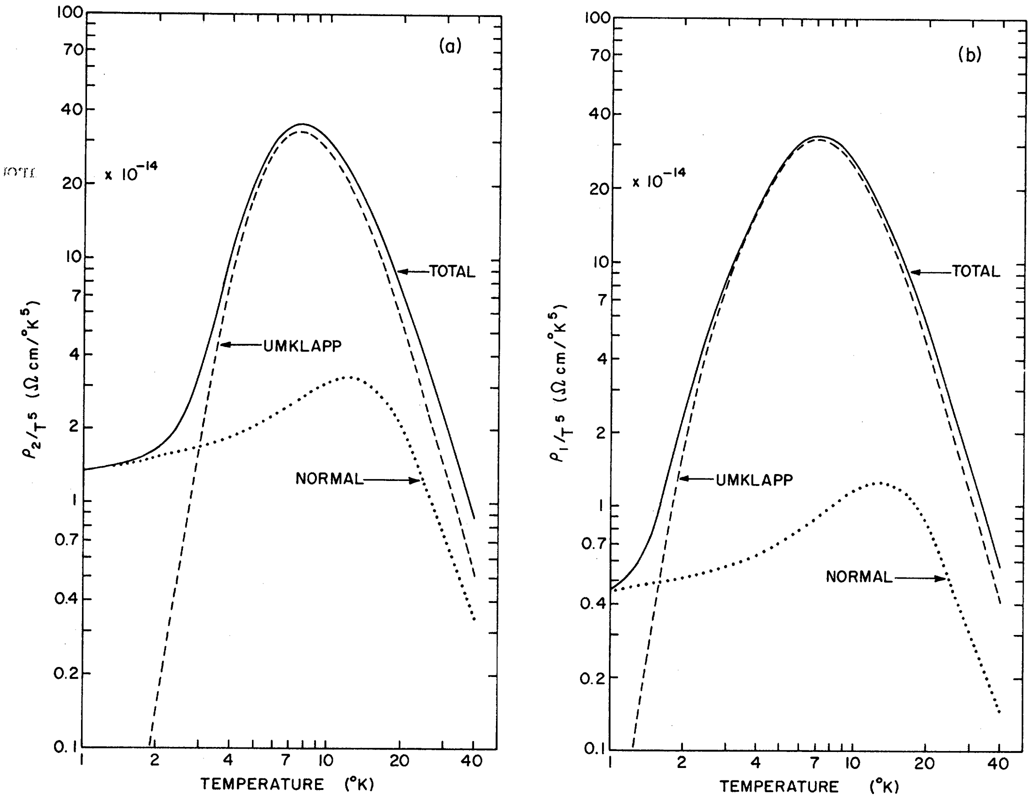

FIG. 5. Relaxation times plotted as the coefficient of the T^5 power of resistivities for the Debye-like phonon model for (a) ultrasonic attenuation and (b) electrical resistivity for the Ashcroft pseudopotential.

model in the calculations, Fig. 5(a) depicts the results for the attenuation and 5(b) the resistivity with the Ashcroft pseudopotential and an isotropic Debye-like phonon model with quadratic energy dispersion and one purely longitudinal branch and two degenerate purely transverse branches.[19] Figure 5 should be compared with Fig. 2. The normal component in Figs. 5(a) and 5(b) is underestimated, but this is not unexpected since it is very sensitive to the elastic constants. In addition, the "transverse" phonon modes in the realistic model are not actually purely transverse throughout the first Brillouin zone; the factor $|\vec{\epsilon}(\vec{q};\lambda)\cdot\vec{q}|^2$ in Eq. (4) for the "transverse" modes is not identically zero throughout the zone and there is a significant contribution to the normal component from transverse modes. The umklapp components in Fig. 5 are in excess by a large amount over those in Fig. 2; this is due to an underestimation of the sound velocity of the transverse modes in the Debye-like model.

IV. CONCLUSION

A thorough analysis of the relative importance of normal and umklapp scattering processes in the low-temperature ultrasonic attenuation due to electron-phonon interactions in the local limit and the ideal electrical resistivity has been presented. Calculations with various pseudopotentials have shown that the umklapp scattering processes play the determining role in the complicated temperature dependence observed in the low-temperature ideal resistivity of Ref. 5. Umklapp scattering processes play a less pronounced role in the ultrasonic attenuation but, nevertheless, the attenuation is enhanced over that due to normal scattering processes alone. In particular, a dependence more rapid than T^5 is found in the region 5–2.5 °K and is attributed mainly to the umklapp processes.

As pointed out by Rice and Sham,[1] there is a disagreement between the measured and calculated values of τ_2/τ_1[23]; this seems to call for further measurements of the ultrasonic attenuation in the local limit since the resistivity results are in excellent accord.

It should be pointed out that phonon-drag effects have not been taken into account in the calculations of the resistivity and that the trial function[15] used is not necessarily the optimum in view of the moderately dominant role played by umklapp processes. Scattering due to umklapp processes alone is expected to be highly anisotropic at low temperatures. Yet the agreement with experiment seems not to be appreciably affected by these approximations at temperatures above 1.5 °K.

ACKNOWLEDGMENTS

We have greatly benefitted from discussions with N. W. Ashcroft, J. W. Wilkins, L. J. Sham, B. G. Nickel, T. M. Rice, and B. W. Maxfield.

---

[*] Work supported by the Advanced Research Projects Agency through the facilities of the Materials Science Center at Cornell University, MSC Report No. 1564.

[†] National Research Council of Canada Postdoctorate Fellow.

[‡] National Science Foundation Trainee.

[1] T. M. Rice and L. J. Sham, Phys. Rev. B 1, 4546 (1970).

[2] G. G. Natale and I. Rudnick, Phys. Rev. 167, 687 (1968).

[3] For example, J. C. Garland and R. Bowers, Phys. Rev. Letters 21, 1007 (1968); V. S. Tsoĭ and V. F. Gantmakher, Zh. Eksperim. i Teor. Fiz. 56, 1232 (1969) [Sov. Phys. JETP 29, 663 (1969)].

[4] S. B. Woods, Can. J. Phys. 34, 223 (1956); see also the further interpretation by M. Kaveh and N. Wiser, Phys. Rev. Letters 26, 635 (1971).

[5] J. W. Ekin, Phys. Rev. Letters 26, 1550 (1971); J. W. Ekin, P. N. Trofimenkoff, and B. W. Maxfield, Bull. Am. Phys. Soc. 16, 344 (1971).

[6] R. Peierls, Ann. Phys. (Paris) 12, 154 (1932); see, for example, R. E. Peierls, *Quantum Theory of Solids* (Oxford U. P., Oxford, England, 1965), p. 135; J. M. Ziman, *Electrons and Phonons* (Oxford U. P., Oxford, England, 1960), p. 369.

[7] J. M. Ziman, Proc. Roy. Soc. (London) A226, 436 (1954); M. Bailyn and H. Brooks, Bull. Am. Phys. Soc. 1, 300 (1956); M. Bailyn, Phys. Rev. 120, 381 (1960); J. G. Collins and J. M. Ziman, Proc. Roy. Soc. (London) A264, 60 (1961); A. Hasegawa, J. Phys. Soc. Japan 19, 504 (1964); T. M. Rice and L. J. Sham, Ref. 1.

[8] H. E. Bömmel, Phys. Rev. 96, 220 (1954).

[9] L. Mackinnon, Phys. Rev. 98, 1181 (1955); H. E. Bömmel, *ibid.* 100, 758 (1955).

[10] W. P. Mason, Phys. Rev. 97, 557 (1955); R. W. Morse, *ibid.* 97, 1716 (1955); C. Kittel, *ibid.* 98, 1181 (1955); Acta Met. 3, 295 (1955); A. B. Pippard, Phil. Mag. 46, 1104 (1955); M. S. Steinberg, Phys. Rev. 111, 425 (1958); T. Holstein, *ibid.* 113, 479 (1959).

[11] A. Akhiezer, J. Phys. (USSR) 1, 289 (1939).

[12] For both the attenuation and resistivity, many-body and band corrections cancel out in the final results [see Ref. 1 and T. M. Rice, Phys. Rev. 175, 858 (1968)]. Thus the free-electron values will be used throughout this paper.

[13] A. B. Bhatia and R. A. Moore, Phys. Rev. 121, 1075 (1961).

[14] D. Shoenberg and P. J. Stiles, Proc. Roy. Soc. (London) A281, 62 (1964).

[15] The expression in Eq. (4) for the electrical resistivity can be obtained from the Kohler variational expression with a trial function proportional to $\vec{k}\cdot\hat{u}$, where $\vec{k}$ is the electron wave vector and $\hat{u}$ is a unit vector in the direction of the applied electric field. See M. Kohler, Z. Physik 124, 772 (1948); 125, 679 (1949); E. H. Sondheimer, Proc. Roy. Soc. (London) A203, 75 (1950); J. M. Ziman, Ref. 6, p. 275.

[16] R. A. Cowley, A. D. B. Woods, and G. Dolling, Phys. Rev. 150, 487 (1966); the sound velocity data used in this fit are those of W. R. Marquardt and J. Trivisonno, J. Phys. Chem. Solids 26, 273 (1965).

[17] G. Gilat and L. J. Raubenheimer, Phys. Rev. 144, 390 (1966); see also B. Hayman and J. P. Carbotte, Can. J. Phys. (to be published).

[18] C. S. Barrett, Acta Cryst. 9, 671 (1956).

[19] P. B. Allen and M. L. Cohen, Phys. Rev. 187, 525 (1969).

[20] J. Bardeen, Phys. Rev. 52, 688 (1937); apart from the lattice parameter, the parameters in this model are those given by A. Hasegawa, Ref. 7. The total electrical resistivity results calculated with this pseudopotential are in good agreement with the previously tabulated results of Hasegawa.

[21] N. W. Ashcroft, Phys. Letters 23, 48 (1966); $R_c = 1.13$ Å.

[22] M. J. G. Lee and L. M. Falicov, Proc. Roy. Soc. (London) A304, 319 (1968); for details see Ref. 1. We are grateful to L. J. Sham for providing a tabulation of these two pseudopotentials.

[23] Although the ratios τ_2/τ_1 calculated from this work are in excellent agreement with those of Ref. 1, there is a discrepancy in the absolute values when compared individually. This is attributed to an inadvertent constant numerical error in evaluating the resistivities quoted in Ref. 1, T. M. Rice and L. J. Sham, Phys. Rev. B 4, 674 (1971).