

Ultrasonic Attenuation by Conduction Electrons in Potassium*

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 (Received 30 October 1967)

Measurements of ultrasonic attenuation due to electron-lattice interaction were made on potassium wire samples with a pulse-echo or resonant-decay technique from 2.5 to 20°K at frequencies between 200 and 400 kc/sec ($ql \ll 1$ region). The magnitude of the attenuation was found to be on the average 1.6 times larger than predicted by the free-electron theories on the basis of the measured values of electrical conductivity. The imperfect knowledge of the background introduced a maximum 30% error. Various reasons for the observed discrepancy are discussed, but the results may be within the limits of the approximations involved in the theories. The temperature and frequency dependence of the attenuation were found to be in good agreement with the theoretical expectations.

INTRODUCTION

FOLLOWING Bömmel's^{1,2} and MacKinnon's³ experiments on the attenuation of a sound wave in metals by interaction with the conduction electrons, numerous measurements of the effect were made in several metals.^{4,5} At the same time a number of theoretical explanations⁶ were formulated, in addition to the original treatment given by Akhiezer.⁷ These theories involve the assumption of a spherical Fermi surface and on this basis it is found that in the region of $ql \ll 1$ (q is the wave propagation constant and l is the electronic mean free path) the attenuation constant A of a longitudinal wave propagating in an infinite medium can be expressed by

$$A = (2/15)(Nmv_F^2\omega^2\tau/\rho v_s^3), \quad (1)$$

where N is the number of free electrons per unit volume, m is the electronic mass, v_F is the Fermi velocity, ρ is the density, v_s is the ultrasonic velocity, ω is the angular frequency of the wave, and τ is the relaxation time, generally assumed to be equal to the one which appears in the expression for the electrical conductivity. Similar relations are found for the range of $ql \gg 1$ and for the case of transverse waves.

The experimental results to date generally confirm the frequency and temperature dependence predicted by these theories, but the absolute magnitude of the

attenuation has been found to exceed the calculated value. In view of the fact that the measurements were made on nonideal metals, i.e., nonspherical Fermi surfaces, it seems possible that part or all of the discrepancy could be attributed to this fact. Some theories were developed for nonideal metals,⁸ but in their results the amplitude of the attenuation depends on certain factors which are not well known and cannot be accurately calculated for each specific case.

For these reasons, it appears that measurements on alkali metals would be of interest. Stern *et al.*⁹ attempted such measurements in sodium, but an accurate determination of the electron-lattice component of the attenuation was prevented by the presence of a large and strongly temperature-dependent acoustic loss associated with the martensitic transformation. The next best approximation to an ideal metal is potassium, where no phase change has been observed. Most theoretical and experimental studies of the Fermi surface of potassium indicate that it is spherical within 1%.

EXPERIMENTAL

The potassium samples were prepared from metal of 99.98% nominal purity obtained from MSA Research Corporation. Most of the experimental difficulties introduced by the mechanical and chemical properties of potassium were overcome by extruding the samples in the form of wires (~ 0.2 cm diam, ~ 10 cm long) at liquid-nitrogen temperature and by keeping them thereafter at this or a lower temperature. However, the primary reason for this procedure was to avoid an excessive growth of the crystal grain size, thereby limiting the acoustic losses due to thermoelastic relaxation and grain-boundary scattering; the recrystallization temperature of potassium was found to be near 105°K. The physical characteristics of the samples used in the

* This work was supported by the U. S. Office of Naval Research.

¹ H. E. Bömmel, Phys. Rev. **96**, 220 (1954).

² H. E. Bömmel, Phys. Rev. **100**, 758 (1955).

³ L. Mackinnon, Phys. Rev. **98**, 1210 (1955).

⁴ W. P. Mason and H. E. Bömmel, J. Acoust. Soc. Am. **28**, 930 (1956); D. H. Filson and E. Lax, in *Proceedings of the Third International Congress on Acoustics, Stuttgart, 1959* (Elsevier Publishing Co., New York, 1961); E. Lax, Phys. Rev. **115**, 6, 1591 (1959); R. E. Macfarlane, J. A. Rayne, and C. K. Jones, Phys. Letters **19**, 354 (1965); R. J. Kolouch and K. A. McCarthy, Phys. Rev. **139**, A700 (1965); E. S. Bliss and J. A. Rayne, Phys. Letters **23**, 38 (1966). For additional references (experimental and theoretical) see the articles by H. Stoltz, Phys. Status Solidi **3**, 1153 (1963), and by N. Tepley, Proc. IEEE **53**, 1586 (1965).

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⁷ A. Akhiezer, J. Phys. (USSR) **1**, 290 (1939).

⁸ E. I. Blount, Phys. Rev. **114**, 418 (1959); A. B. Pippard, Proc. Roy. Soc. (London) **257**, A165 (1960); R. W. Morse, *Progress in Cryogenics* (Heywood and Company Ltd., London, 1959), Vol. I, p. 226; R. G. Chambers, in *Proceedings of the Seventh International Conference on Low Temperature Physics* (The University of Toronto Press, Toronto, 1961), p. 243.

⁹ R. Stern, G. G. Natale, and I. Rudnick, J. Phys. Chem. Solids **27**, 9 (1966).

TABLE I. Characteristics of the potassium samples.

Sample	Length (cm)	Diameter (cm)	$\rho_{273^\circ\text{K}}/\rho_{4,2^\circ\text{K}}^a$	Anneals
K1	10.4	0.208	1790	none
K10	10.9	0.208	1708	1 h at 105°K
K11	10.3	0.208	10	none
K12	9.8	0.208	2440	1 h at 110°K
K13	9.6	0.208	1342	none
K18	10.0	0.208	1187	none
K19	10.0	0.208	1276	none

^a $\rho_{273^\circ\text{K}} = 6.10 \times 10^{-6} \Omega \text{ cm}$ [L. Hackspill, Compt. Rend. 151, 305 (1910)].

present experiments are shown in Table I. A single PZT transducer was bonded to the samples by using isopentane, which was found to be the best among the few compounds which can be used to make an acoustic bond at temperatures near liquid nitrogen.¹⁰

The acoustic measurements were made with a pulse-echo technique or a resonant-decay method; apart from minor modifications, the experimental apparatus used has been described in a previous paper.⁹ In the frequency range used (approximately 200 to 400 kc/sec) the longitudinal waves generated in the samples were Young's modulus waves. The measurements were made at temperatures between 2.5 and 77°K, but the electron-lattice component of the attenuation becomes important only below about 20°K. Measurements of electrical resistivity as a function of temperature, necessary to determine the relaxation time τ appearing in Eq. (1), were made with a four-lead potentiometer method between 2.5 and 20°K.

RESULTS

Amplitude of the Effect

In addition to the electron-lattice component of the attenuation, there are present other losses (the "background") due to the transducer-bond-support system, the polycrystalline nature of the sample and various effects associated with the motion of dislocations. The magnitude and temperature dependence of most of these losses were determined by measuring the attenuation in a sample doped with about 1.5% of sodium. The added impurity had the effect of drastically decreasing the electronic mean free path, thereby essentially eliminating the electron-lattice component of the attenuation; in addition, however, the impurity provided pinning points for the dislocations, thus modifying the effective length of the dislocation loops. There is, therefore, an uncertainty about the temperature dependence of the dislocation component of the attenuation which in turn introduces a maximum error in the electron-lattice interaction data estimated to be between 15% and 30% (depending on the frequency) at the lowest temperature.

¹⁰ R. Stern, G. G. Natale, and I. Rudnick, Rev. Sci. Instr. 35, 1077 (1964).

Figure 1 shows the electron-lattice attenuation as a function of temperature at 219 kc/sec; these data were obtained by subtracting the background measured at about 20°K from the total attenuation and correcting for the temperature dependence obtained in the impure sample mentioned above. The data represent the average of five runs using four different samples; the results were reproducible within 10%.

The theoretical curve has been calculated from the expression⁵

$$A = (2/15)(f^2/\rho v_y^3)(1+\nu^2)(h^2/e^2)(3\pi^2 N)^{2/3}\sigma,$$

where v_y is the velocity of a Young's modulus wave, ν is Poisson's ratio, σ is the electrical conductivity, f is the ultrasonic frequency, e is the electronic charge, and h is Planck's constant.

This relation is valid for a Young's modulus wave, and it differs from the free medium expression by the factor $(1+\nu)^2$, and by the fact that v_y replaces the velocity for longitudinal waves in an infinite medium; in addition, the relaxation time has been expressed in terms of the electrical conductivity. It is apparent that if the theoretical curve is multiplied by the factor 1.7, it coincides with the measured curve within the experimental error (estimated at 10%).

Similar results were obtained at 277 and 368 kc/sec, as shown in Figs. 2 and 3. The average value of the excess factor over the frequency range investigated is about 1.6.

Temperature Dependence

As pointed out above, the temperature dependence of the attenuation appears to be the same as that of the electrical conductivity. Measurements of electrical re-

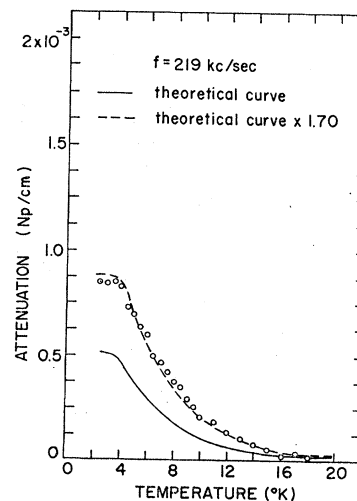


FIG. 1. Temperature dependence of average electron-lattice attenuation in potassium at 219 kc/sec. The data points are corrected for background losses as described in the text. The theoretical curve is based on measurements of electrical resistivity.

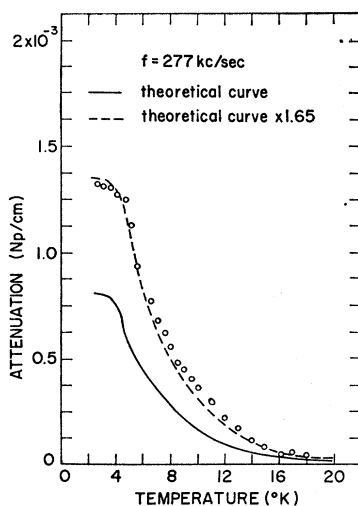


FIG. 2. Temperature dependence of average electron-lattice attenuation in potassium at 277 kc/sec. The data points are corrected for background losses as described in the text. The theoretical curve is based on measurements of electrical resistivity.

sistivity versus temperature made on two samples in different states of anneal are shown in Fig. 4. The ideal resistivity can be obtained by subtracting the residual resistivity from the total values, on the basis of Matthiessen's rule; this has been done in Fig. 5. It appears that a T^5 law with a proportionality coefficient of 2.6×10^{-3} is followed below approximately 7°K ; at higher temperatures the dependence decreases and becomes about T^3 at 20°K . These results are in fair agreement with the Bloch-Grüneisen theory which predicts a T^5 dependence below approximately $\frac{1}{10}$ of a characteristic temperature which for potassium is close

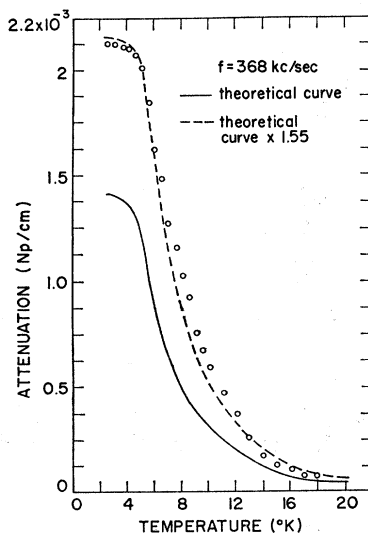


FIG. 3. Temperature dependence of average electron-lattice attenuation in potassium at 368 kc/sec. The data points are corrected for background losses as described in the text. The theoretical curve is based on measurements of electrical resistivity.

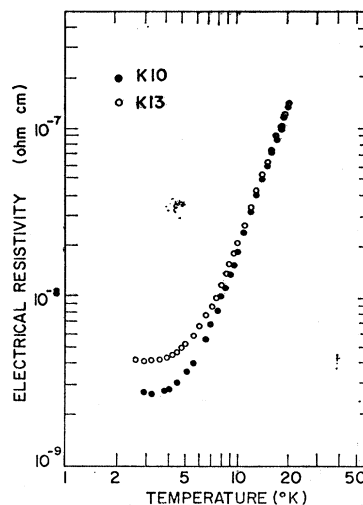


FIG. 4. Electrical resistivity versus temperature in sample K13 (unannealed) and in sample K10 (annealed one hour at 105°K). The measurements were made with a four lead potentiometer method.

to 90°K . A very recent calculation has been made by Hasegawa¹¹ assuming a spherical Fermi surface and using a phonon spectrum obtained from measurements of specific heat and neutron spectrometry rather than from an idealized model; our resistivity data are given together with the Hasegawa curve in Fig. 6 and the agreement is good, with a possible 5% discrepancy at the lowest temperature, where, however, the experimental error is largest.

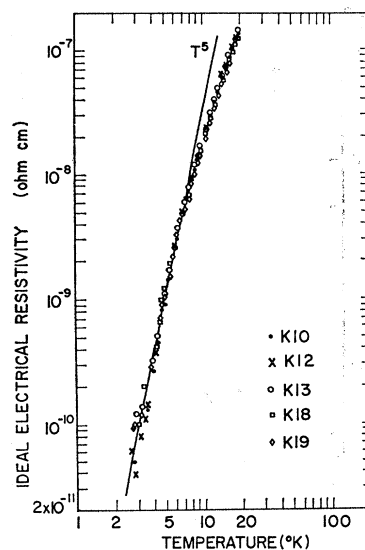


FIG. 5. Temperature dependence of the ideal electrical resistivity in various potassium samples. The T^5 curve is shown for comparison.

¹¹ A. Hasegawa, J. Phys. Soc. Japan 19, 504 (1964).

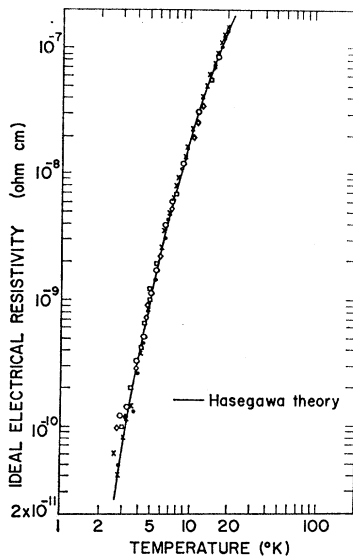


FIG. 6. Comparison of experimental data on ideal electrical resistivity in potassium with the Hasegawa theory (see Ref. 11).

Frequency Dependence

Measurements of attenuation as a function of frequency (approximately 200 to 400 kc/sec) were made at 4.2°K. The total attenuation follows an $f^{1.6}$ law, but when the background present at about 18°K, which is proportional to $f^{1.26}$, is subtracted from the total attenuation, the dependence of the difference becomes $f^{1.95}$; this is illustrated in Fig. 7. The value of ql for the frequency range used in these measurements varies

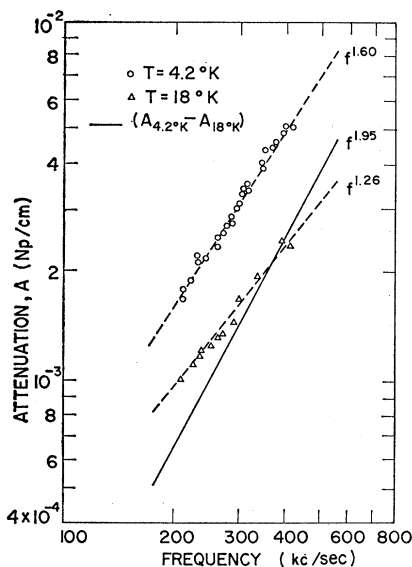


FIG. 7. Frequency dependence of the attenuation in potassium at 4.2 and 18°K, and frequency dependence of their difference. The difference represents primarily the electron-lattice component of the attenuation.

between 3.5×10^{-2} and 7×10^{-2} , and in this region the theory predicts a square law dependence. A slightly lower value of the exponent can be probably attributed to a small residual background loss present at 4.2°K.

DISCUSSION AND CONCLUSIONS

The present experiments seem to establish that even in potassium the measured electron-lattice attenuation exceeds the value predicted by the free-electron theories by an average factor of 1.6 over the frequency range investigated.

It should be noted that the calculated value of the attenuation has an uncertainty estimated at about 25%. This arises from the probable errors contained in the values of some factors which appear in the expression used for the calculation. The largest uncertainty is probably the value of Poisson's ratio which was calculated from the single-crystal elastic constants of Marquardt and Trivisonno¹² using the method of Kröner¹³; the value of ν thus obtained (0.350) has an estimated probable error of 8%. The values of v_ν , which were both measured from the transit time of the pulses and calculated from the single-crystal elastic constants, are probably correct to within 3%; a similar error is present in the measured values of σ . However, part of the discrepancy could be attributed to the fact that the relaxation time for the acoustic-attenuation process is not necessarily the same as the one for the electrical resistivity. A calculation by Bhatia and More¹⁴ indicates that the difference between the relaxation times could be as much as 20% and, if umklapp processes are taken into account, the relaxation time for the acoustic attenuation would be the larger of the two. This would account for about $\frac{1}{3}$ of the observed discrepancy in the present case. In addition, it must be recognized that even small deviations from a spherical Fermi surface, as present in potassium, are likely to introduce a sizable excess attenuation.

It might well be that the observed difference between measured and calculated values is within the limits of the approximations involved in the theories for both ideal and nonideal metals. If this is the case, a closer agreement cannot be expected until some of these approximations are improved upon, and more refined calculations can be made to take into account the effect of slight departures from the ideal model.

ACKNOWLEDGMENT

The authors gratefully acknowledge the help given by Taylor G. J. Wang during the course of the experiments.

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¹³ E. Kröner, *Z. Physik.* **151**, 504 (1958).

¹⁴ A. B. Bhatia and R. A. Moore, *Phys. Rev.* **121**, 1075 (1961).