# Ultrasonic Attenuation of Transverse Waves in V, Nb, and Ta for $ql<1$*†

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The temperature dependence of the ultrasonic attenuation coefficient of transverse waves with a wave vector parallel to ⟨110⟩ was measured in superconducting V, Nb, and Ta. The product of the wave vector times the mean free path $ql$ was smaller than one during these measurements. The sharp drop in the attenuation coefficient below the transition temperature, which is found when $ql>1$, was not present. The zero-temperature energy gap was found to be $3.6\pm0.2\ kT_c$ for V, $3.5\pm0.3kT_c$ for Nb, and $3.5\pm0.2kT_c$ for Ta.

## INTRODUCTION

FOR large values of $ql$, where $q$ is the propagation vector and $l$ is the electron-phonon mean free path, ultrasonic attenuation of transverse waves experiences a sharp drop when the temperature is lowered below the transition temperature $T_c$ of the superconductor.[1] Morse[1] and Tsuneto[2] attribute this sharp drop to the fact that the onset of the Meissner effect screens the transverse magnetic field proposed by Pippard[3] to account for ultrasonic attenuation of shear waves in the normal state. Morse and Claiborne[4] have proposed that the sharp drop in attenuation should vanish for $ql\ll1$. In this range[5] the ratio of the attenuation coefficient in the superconducting and normal state is given by

$$\alpha_s/\alpha_n=2/(\epsilon^{\epsilon_0/kT}+1), \quad (1)$$

where $\alpha$ is the attenuation coefficient, $s$ refers to the superconducting state, $n$ to the normal state, $2\epsilon_0$ is the temperature-dependent superconducting energy gap, $T$ is the absolute temperature, and $k$ is Boltzmann's constant. This relationship was first derived by Bardeen, Cooper, and Schrieffer[6] (BCS) for longitudinal waves for $ql\gg1$.

The temperature dependence of the attenuation coefficient of transverse waves was measured in superconducting vanadium, niobium, and tantalum. In all these experiments the $ql$ product was small. Particular attention was paid to the possible existence of a sharp drop of $\alpha_s/\alpha_n$ near $T_c$. None was found.

The ultrasonic and cryogenic equipment and the pulse echo technique employed for making the measurements are described elsewhere.[7] A 15-Mc/sec $AT$ quartz crystal was used as the transducer. It was cemented[8] to the end of the samples with an epoxy resin. This was the only procedure which gave successful bonds between the $AT$ crystals and our samples. Detailed dilatational measurements have already been reported on the vanadium sample[7] and the tantalum sample.[9] Only gross longitudinal measurements were done on the niobium sample since the total attenuation change which could be observed for this sample was too small to be measured accurately.

The properties of the samples are tabulated in Table I. The vanadium and tantalum sample numbers, 3 and 2, respectively, are those used in reporting the results on dilatational waves.

## VANADIUM

Measurements were made on a pure vanadium sample at 135 Mc/sec with the polarization vector along ⟨110⟩, Fig. 1. The ratio of the attenuation in the superconducting state to that in the normal conducting state is plotted as a function of the reduced temperature $T/T_c$. No sharp drop is present near the transition temperature. The dots in these figures indicate points obtained while the temperature of each of the samples was increased in successive steps of about 0.03°K. The crosses indicate data taken while the temperature was being decreased. $L$ is the distance travelled by the particular pulse being measured and therefore $a_nL$ is the total attenuation of the pulse in the normal state. The solid line is plotted according to Eq. (1) with a zero temperature energy gap of $3.5\ kT_c$. The small oscillatory deviations of the experimental data from the theoretical curve are discussed in Ref. 7 and are believed to be an interference of the high-order elastic wave modes which comprise the pulse. This set of shear measurements was performed while an $X$-cut quartz crystal was still bonded to the opposite end of the sample. The possibility was being checked, of bonding an $X$-cut and an $AT$-cut quartz crystal to opposite ends of a sample in order to save the time required to remove the crystals whenever a different ultrasonic mode was being investigated. This procedure proved to have the disadvantage that the $X$-cut crystal produced enough rf signal to interfere with that coming from the shear

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[1] See for instance R. W. Morse, *Progress in Cryogenics* (Heywood and Company Ltd., London, 1959), Vol. I, pp. 219 ff.
[2] T. Tsuneto, Phys. Rev. **121**, 402 (1961).
[3] A. B. Pippard, Phil. Mag. **46**, 1104 (1955).
[4] R. W. Morse, IBM J. Res. Develop. **6**, 58 (1962).
[5] M. Levy, Phys. Rev. **131**, 1497 (1963).
[6] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[7] J. L. Brewster, M. Levy, and I. Rudnick, Phys. Rev. **132**, 1062 (1963).
[8] M. Levy and I. Rudnick. J. Acoust. Soc. Am. **34**, 520 (1962).
[9] M. Levy and I. Rudnick, Phys. Rev. **132**, 1062 (1963).

TABLE I. Properties of the samples.

| | V No.3 | Nb | Ta No. 2 |
|---|---|---|---|
| Transtion temperature[a] | 5.28°K | 8.89°K | 4.42°K |
| Resistivity ratio[b] | 170 | 8 | 40 |
| Diamond Pyramid Hardness[c] | 58 | 190 | 86 |
| Purity[d] | 99.95% | 99.7% | 99.9% |
| Size (diameter and length)[e] | 0.25×0.4908 in. | 0.25×1.045 in. | 0.25×1.0963 in. |
| Axial orientation[f] | ⟨110⟩ | ⟨110⟩ | ⟨110⟩ |
| Method of growth[g] | electron beam zone refined | | electron beam zone refined |
| Source | Atomics Internat. and Iowa State University | Linde | M.R.C. |

[a] The transition temperature was determined acoustically.
[b] The residual resistivity ratio was measured by an eddy current technique. [C. P. Bean, R. W. DuBlois, and L. B. Nesbitt, J. Appl. Phys. **30**, 1976 (1959); see also R. Stern, M. Levy, R. Kagiwada, and I. Rudnick. Appl. Phys. Letters **2**, 80 (1963)].
[c] A Tukon Tester was used in making these measurements.
[d] The purity of the vanadium sample was provided by O. N. Carlson and C. V. Owen. See O. N. Carlson and C. V. Owen, J. Electrochem. Soc. **108**, 88 (1961). The purity of the niobium sample was obtained from Linde, that of the tantalum sample from M.R.C.
[e] The ends of the cylinder were optically polished and ground parallel to 0.1 mils.
[f] The crystals were grown along ⟨110⟩.
[g] Quarter-inch cylinders of vanadium and tantalum were grown which were then cut to the proper length. The niobium sample was centerless ground to the proper dimensions by Linde.

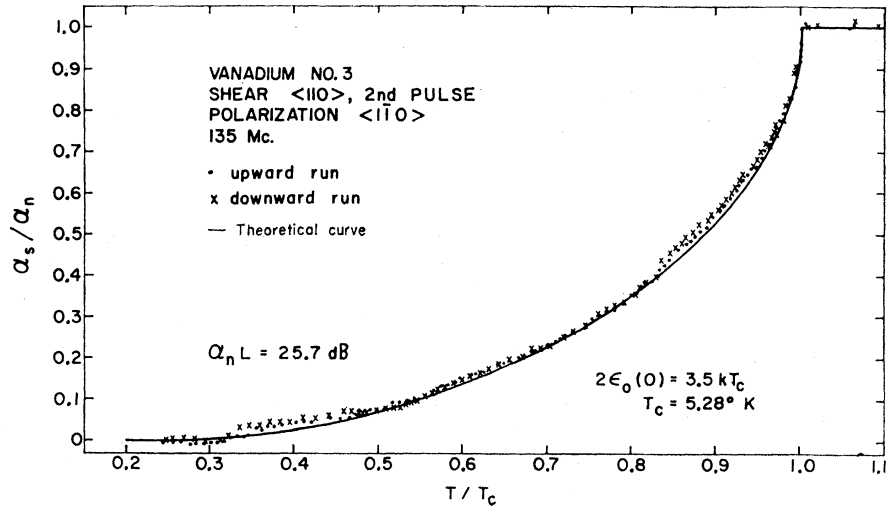


FIG. 1. Normalized attenuation of transverse waves as a function of reduced temperature in V at 135 Mc/sec.

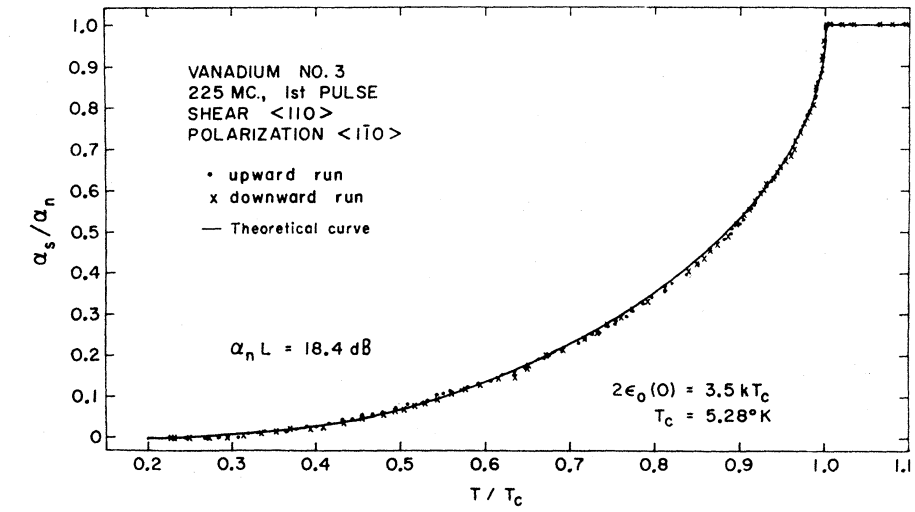


FIG. 2. Normalized attenuation of transverse waves as a function of reduced temperature in V at 225 Mc/sec.

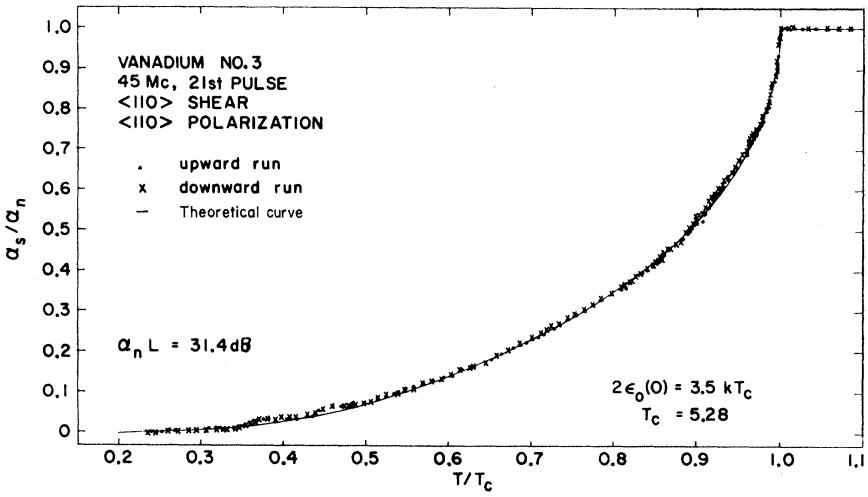


FIG. 3. Normalized attenuation of transverse waves as a function of reduced temperature in V at 45 Mc/sec.

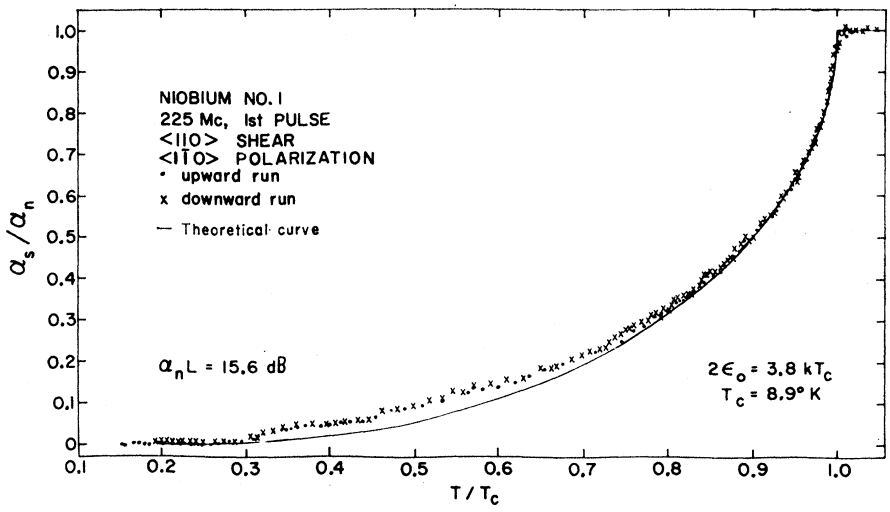


FIG. 4. Normalized attenuation of transverse waves as a function of reduced temperature in Nb at 225 Mc/sec.

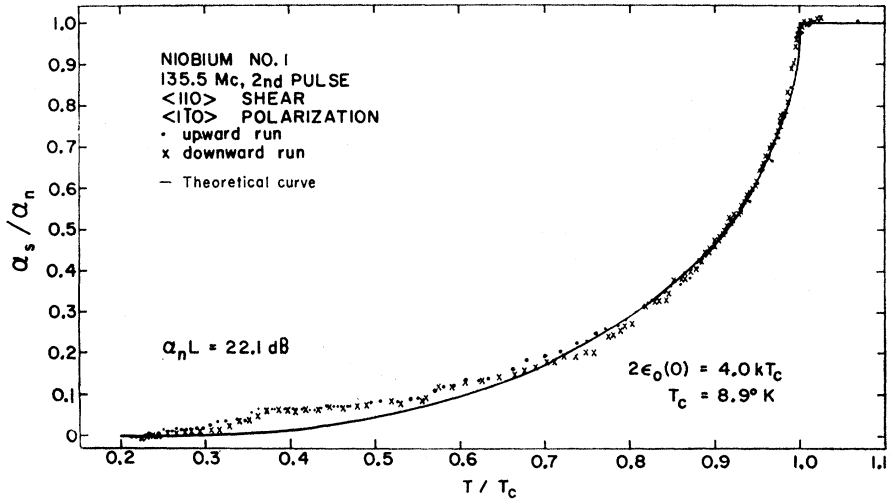


FIG. 5. Normalized attenuation of transverse waves as a function of reduced temperature in Nb at 135 Mc/sec.

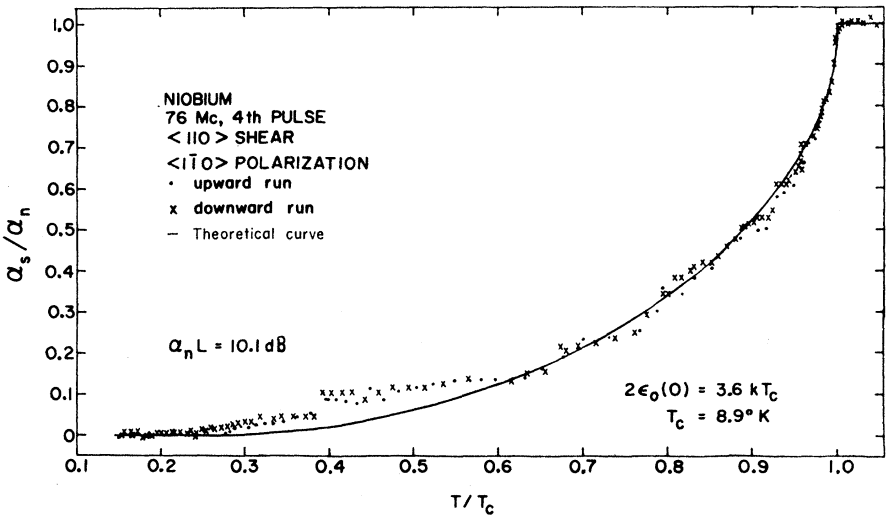


FIG. 6. Normalized attenuation of transverse waves as a function of reduced temperature in Nb at 76 Mc/sec.

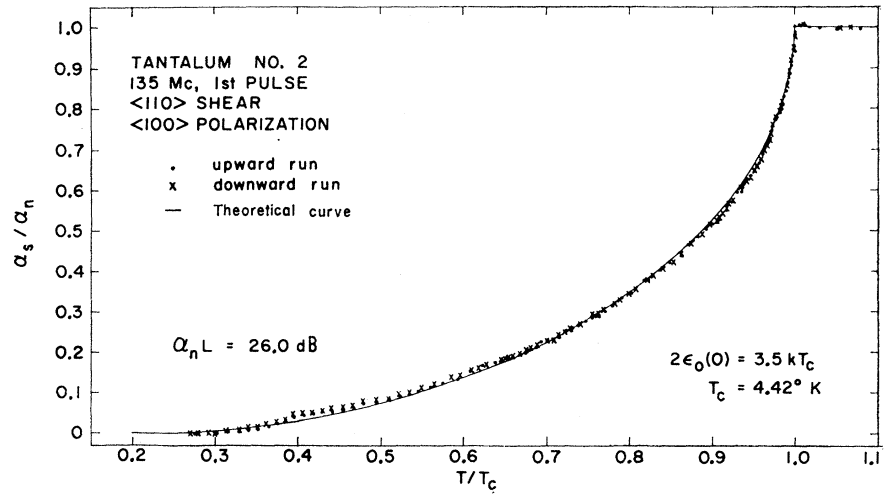


FIG. 7. Normalized attenuation of transverse waves as a function of reduced temperautre in Ta at 135 Mc/sec.

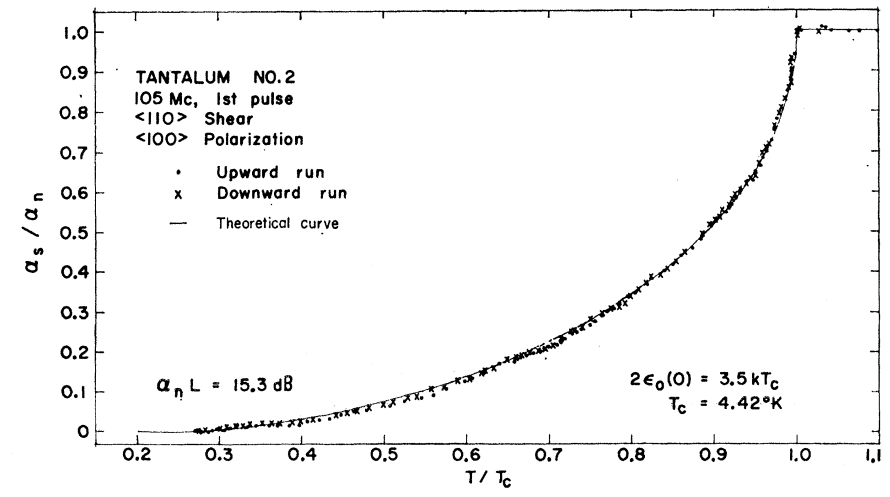


FIG. 8. Normalized attenuation of transverse waves as a function of reduced temperature in Ta at 105 Mc/sec.

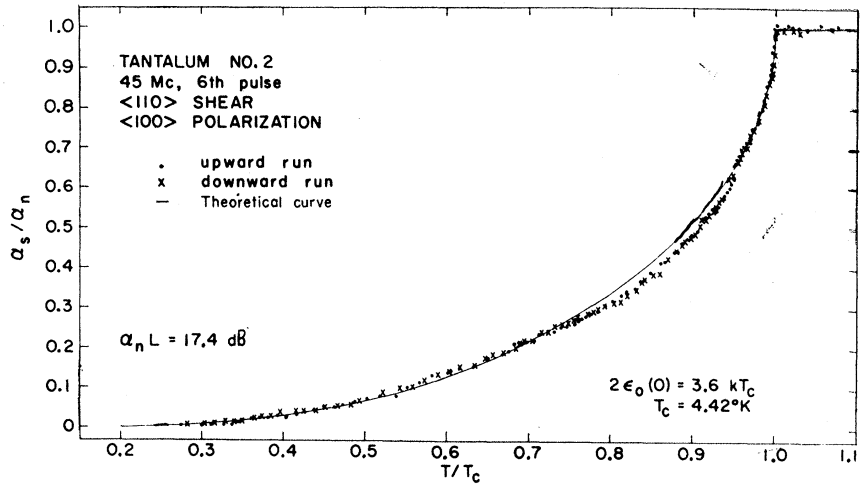


FIG. 9. Normalized attenuation of transverse waves as a function of reduced temperature in Ta at 45 Mc/sec.

crystal. Therefore, in order to ensure the validity of our data, another set of experiments was performed after both crystals had been removed and a properly aligned $AT$-cut crystal was bonded to the vanadium sample. Figure 2 shows our results at 225 Mc/sec with the polarization vector along ⟨110⟩. There is no noticeable change between the two sets of data. Again no marked drop in $\alpha_s/\alpha_n$ was observed near the transition temperature. It is also absent in the data taken at 45 Mc/sec, Fig. 3.

## NIOBIUM

Ultrasonic attenuation of shear waves was investigated at three frequencies: 225 Mc/sec, Fig. 4; 135.5 Mc/sec, Fig. 5; and 76 Mc/sec, Fig. 6. The solid lines are plots of Eq. (1). No sharp drop is apparent near the transition temperature. In this instance it may be argued that since the niobium is relatively impure, it will not have a sharp transition temperature and thus the appearance of a sharp drop will be inhibited. This argument loses its force as it applies to the vanadium and tantalum data since their transition temperatures (as indicated by dilatational wave measurements) were quite sharp. As may be seen in the graphs, the data are fit by theoretical curves with zero-temperature energy gaps of 3.5 $kT_c$, 4.0 $kT_c$, and 3.6 $kT_c$. The high-frequency data are the most reliable.

## TANTALUM

Transverse wave measurements were obtained at three frequencies on the tantalum crystal: 135 Mc/sec, Fig. 7, 105 Mc/sec, Fig. 8, and 45 Mc/sec, Fig. 9. The polarization vector was along ⟨100⟩, and the propagation vector along ⟨110⟩. The solid curves are plotted for zero-temperature energy gaps of 3.5 $kT_c$, 3.5$kT_c$, and 3.6 $kT_c$, respectively. Again there is no evidence of a sharp drop near the transition temperature, and Eq. (1) appears to fit the data fairly well.

## DISCUSSION AND CONCLUSION

The frequency dependence of the attenuation coefficients for shear waves of the three samples is shown in Fig. 10. We have also included values for the attenuation coefficient of longitudinal waves of the three samples. Pippard[3] has shown that for $ql \ll 1$ $\alpha_n$ is proportional to the square of the frequency for both longitudinal and transverse waves. It is evident that this frequency dependence is observed. Pippard also finds, for $ql \ll 1$, that $\alpha_{nt}/\alpha_{nl} = \frac{3}{4}(v_l/v_t)^3$ where $v$ is the

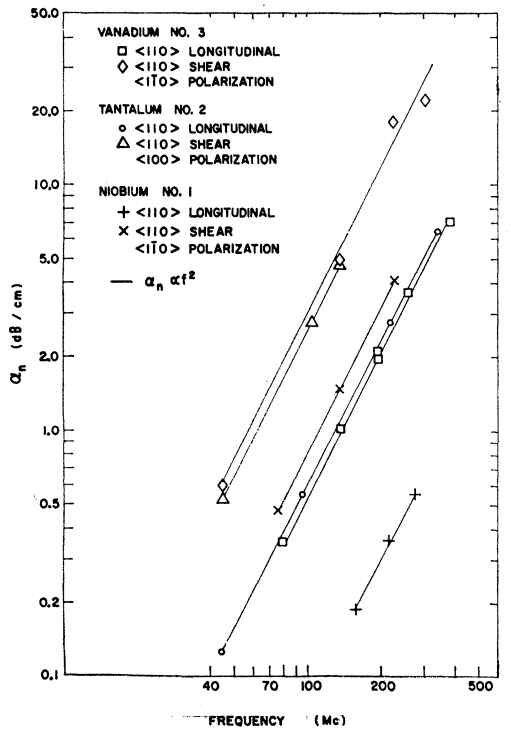


FIG. 10. Frequency dependence of both longitudinal and transverse wave ultrasonic attenuation coefficient in the normal state for V, Nb, and Ta samples.

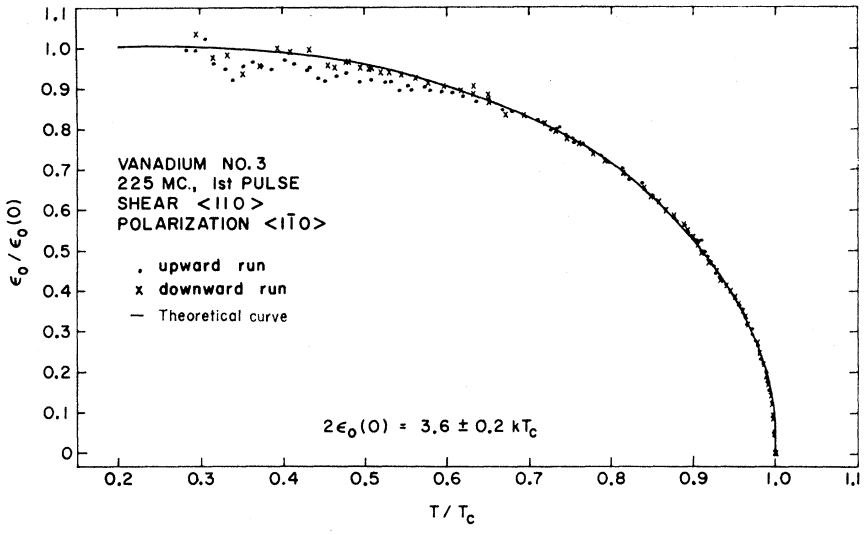


FIG. 11. Temperature dependence of the normalized superconducting energy gap in V.

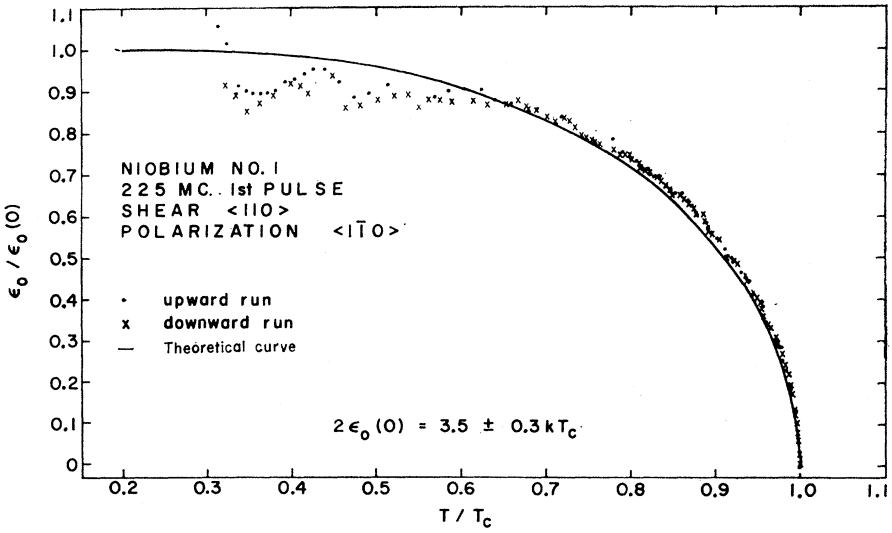


FIG. 12. Temperature dependence of the normalized superconducting energy gap in Nb.

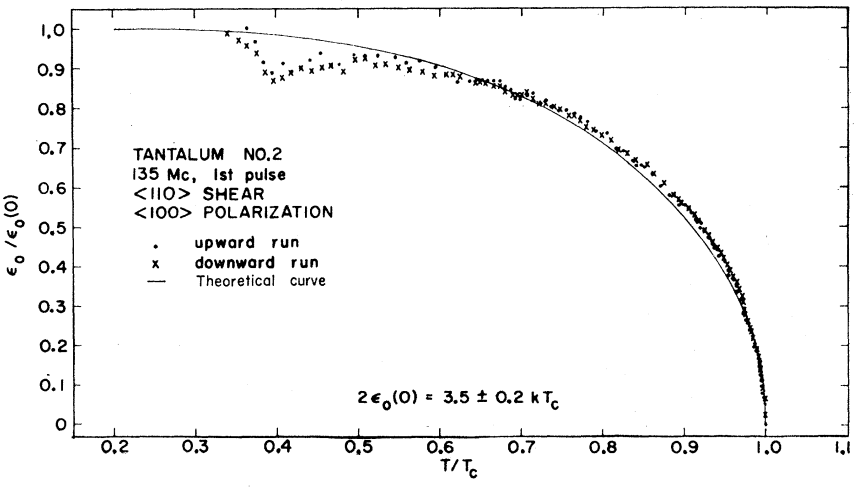


FIG. 13. Temperature dependence of the normalized superconducting energy gap in Ta.

TABLE II. Comparison of experimental and theoretical values of transverse and longitudinal attenuation coefficients due to electron-phonon interaction in the normal state.

| | $\left(\frac{\alpha_{nt}}{\alpha_{nl}}\right)_{\text{exptl.}}$ | $\left(\frac{\alpha_{nt}}{\alpha_{nl}}\right)_{\text{theor.}}$ |
|---|---|---|
| Vanadium | 5.8 | 5.75 |
| Niobium | 6.96 | 5.38 |
| Tantalum | 4.3 | 5.02 |

sound velocity, and the subscripts $t$ and $l$ stand for transverse and longitudinal waves, respectively. Table II compares our experimental results for the ratio with those obtained from the above equation. The values for the sound velocities in Nb and V were obtained from Alers and Waldorf,[10] for Ta from Featherston and Neighbours.[11] There appears to be good agreement between the experimental and theoretical values for V. The longitudinal attenuation measurements in niobium were not too reliable, which may account for the observed discrepancy in this case. However, the difference for tantalum cannot be similarly accounted for, since the total attenuation measured was large enough to give the ratio to an accuracy of at least 5%. This discrepancy between calculated and observed $\alpha_{nt}/\alpha_{nl}$ has also been observed for other metals when $ql>1$.[1] It is difficult to envision how one could be sampling a different portion of the Fermi surface by merely changing the polarization vector of the lattice motion (especially for $ql<1$). Pippard suggests that if different Brillouin zones are involved in the attenuation then the energy levels of two zones may be differently affected by the uniform compression accompanying a longitudinal wave, while this effect would not occur for transverse waves. Thus $(\alpha_{nt}/\alpha_{nl})_{\text{theor.}}$ would be somewhat modified to take into account the periodic transfer of electrons between zones that would occur for longitudinal waves and not for transverse waves. It is probably best to say that the reason for this discrepancy is not yet understood.

The temperature dependence of the energy gap for vanadium as obtained from the 225-Mc/sec data is plotted in Fig. 11. The scatter of the data at low values of $T/T_c$ is due to the fact that any slight deviation of the experimental points of $\alpha_s/\alpha_n$ from their correct value is reflected by a large deviation of $\epsilon_0(T)/\epsilon_0(0)$ for the lower temperatures. The average zero-temperature energy gap $2\epsilon_0(0)$ and the root mean square deviation as computed from all the points using the theoretical values of $\epsilon_0(T)/\epsilon_0(0)$ are also given in Fig. 11. Similar curves are shown for the 225-Mc/sec niobium data, and the 135-Mc/sec tantalum data in Figs. 12 and 13. In Table III we have compared our values of $2\epsilon_0(0)$ for the three metals with those obtained from longitudinal acoustic measurements on the same samples. We have also included values of $2\epsilon_0(0)$ obtained on other samples of V, Nb, and Ta from tunneling experiments, from infrared absorption measurements, from heat capacity data, from thermal conductivity data, and from acoustic measurements. The values in columns I and II were obtained from the shear measurements, the first from visually fitting the data with the proper theoretical curve, the second by averaging $2\epsilon_0(0)$ as obtained from all the points obtained in a reliable run. Column III has the values obtained from longitudinal acoustic measurements on the same samples.[7,9] As was mentioned before, the total longitudinal attenuation measured in niobium was not sufficient to give an accurate estimate of $2\epsilon_0(0)$. Our estimates of the energy gap seem to be in agreement with those obtained by Giaver[12] from tunneling experiments. Sherrill and Edwards[13] also find the same value for Nb from electron tunneling experiments. Our value for vanadium is also in agreement with that obtained by Richards and Tinkham,[14] from infrared absorption measurements. Their values for niobium and tantalum are a little low. However, they felt that their estimate of the transition temperature of the surface of their samples may have been too high since they could only measure the bulk transition temperature and the surface may have become contaminated during the annealing process so that its transition temperature may have been lower than that of the bulk. The values for vanadium and tantalum obtained by Goodman[14] from heat capacity measurements are also in agreement with our values. However, his value for niobium is slightly higher than ours as is also that reported by Connolly and Mendelsson[15] for Ta from thermal conductivity data. The value for the acoustic measurements on vanadium is an average of the values reported by Brewster, Levy, and Rudnick[7] on two single crystals of vanadium with a resistivity ratio of 12 and 19 and by Bohm and Horwitz[16] on one with a resistivity ratio of 130. These measurements were done with longitudinal waves propagating along ⟨110⟩. Dobbs and Perz[17] measured the ultrasonic attenuation coefficient of longitudinal waves propagating along ⟨100⟩ direction of a single crystal of niobium with a resistivity ratio of about 270. Their value is slightly higher than our value in column II. The value

[10] G. A. Alers and D. L. Waldorf, IBM J. Res. Develop. **6**, 89 (1962).

[11] F. H. Featherston and J. R. Neighbours, Bull. Am. Phys. Soc. **7**, 236 (1962).

[12] I. Giaver, *Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications Ltd., London, 1962).

[13] M. D. Sherrill and H. H. Edwards, Phys. Rev. Letters **6**, 460 (1961).

[14] P. L. Richards and M. Tinkham, Phys. Rev. **119**, 575 (1960).

[15] A. Connolly and K. Mendelssohn, Proc. Roy. Soc. (London) **A366**, 429 (1962).

[16] H. V. Bohm and N. H. Horwitz, *Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications Ltd., London, 1962).

[17] E. R. Dobbs and J. M. Perz, *Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications Ltd., London, 1962).

TABLE III. $2\epsilon_0(0)$ of V, Nb, and Ta in units of $kT_c$.

| | I | II | III | Tunneling | Infrared absorption | Heat capacity | Thermal conductivity | Acoustic |
|---|---|---|---|---|---|---|---|---|
| V | 3.5 | 3.6±0.2 | 3.5±0.1 | 3.4 | 3.4 | 3.5 | | 3.45±0.05 |
| Nb | 3.7±0.3 | 3.5±0.3 | | 3.6 | 2.8±0.3 | 3.9 | 3.5 | 3.8 |
| Ta | 3.5±0.1 | 3.5±0.2 | 3.5±0.2 | 3.5 | 3.0 | 3.6 | 3.8 | 3.4±0.2 |

for tantalum was obtained by Levy and Rudnick[9] from longitudinal measurements along the ⟨110⟩ on a sample with a resistivity ratio of 35. It appears that the value of $2\epsilon_0(0)$ for vanadium centers around 3.5 $kT_c$ for all the methods used to determine it. Except for the infrared and thermal conductivity measurements, the values for tantalum also center around 3.5 $kT_c$. There does appear to be a large scatter of the data in niobium. However, tunneling experiments, thermal conductivity measurements, and what we believe to be our most reliable data indicate a value of about 3.6 $kT_c$. It is interesting to note that all these values are consistent with the BCS prediction that $2\epsilon_0(0)=3.5\ kT_c$ for all superconductors.

In conclusion, we have confirmed the theoretical prediction[4,5] of the absence of a sharp drop in $\alpha_s/\alpha_n$ slightly below $T_c$ for transverse waves when $ql<1$. Eq. (1) for $\alpha_s/\alpha_n$ versus $T$ appears to fit the data fairly well. The temperature dependence of the energy gap for V, Nb, and Ta and their zero-temperature energy gap follow the BCS theory. The ratio of the transverse attenuation coefficient to the longitudinal attenuation coefficient in the normal state is compatible with Pippard's theoretical result for V, but no for Ta where our ratio is about 20% lower than that which theory would predict.

## ACKNOWLEDGMENTS

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