Attenuation of Longitudinal Ultrasound in Superconducting Lead*

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The ultrasonic pulse-echo technique was used to measure the absorption of ultrasound in superconducting and normal single-crystal lead. Longitudinal sound waves were propagated in the [100], [110], and [111] crystallographic directions at frequencies between 10 and 70 Mc/sec. The purpose of this invesitgation was to measure the temperature dependence of the superconducting energy gap as well as to attempt to observe its anisotropy. Sufficiently reliable data at low temperatures to do the latter could not be obtained with the present apparatus. The temperature dependence could be reliably measured above a reduced temperature ($t=T/T_c$) of 0.5, and an average zero-temperature energy gap of $2\Delta(0)=(4.1\pm0.2)kT_c$ was obtained using the BCS temperature dependence for extrapolation. In the course of this investigation, we found the attenuation to be amplitude-dependent in the superconducting state, while at the same temperature in the normal state it was amplitude-independent. The energy gap quoted above was measured at the smallest possible amplitudes. The empirical nature of the amplitude effect and its consequences in the determination of the energy gap in lead are discussed. The possibility is pointed out that disagreements between energy-gap temperature dependences predicted by BCS theory and those derived from ultrasonic measurements in elements other than lead may be due to this effect.

I. INTRODUCTION

THE first important ultrasonic measurement on superconducting lead was the pioneering work of Bömmel[1] in 1954. A rapid drop in the attenuation was observed below the transition temperature but no attempt was made to relate this effect to any theories of superconductivity which then existed. According to Bardeen, Cooper, and Schrieffer (BCS) theory[2] the ratio of electronic attenuation in the superconducting state α_s to that in the normal state α_n is given by

$$\frac{\alpha_s}{\alpha_n}=\frac{2}{e^{\Delta(T)/kT}+1}, \tag{1}$$

where $2\Delta(T)$ is the temperature-dependent energy gap. If Bömmel's data are analyzed using this relation, the zero-temperature energy gap turns out to be $8kT_c$. This is in serious disagreement with the BCS prediction of 3.52 kT_c and with the consensus of experimental values for lead obtained by other techniques of approximately 4.2 kT_c.[3] Anomalous superconducting behavior has also been observed in measurements of infrared absorption,[4] thermal conductivity,[5] and critical field[6] in lead. Therefore, further ultrasonic investigations appeared to be in order.

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[1] H. E. Bömmel, Phys. Rev. **96**, 220 (1954).
[2] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[3] For a compilation of results see E. A. Lynton, *Superconductivity* (Methuen and Company, Ltd, London, 1962), p. 92.
[4] D. M. Ginsberg and M. Tinkham, Phys. Rev. **118**, 990 (1960).
[5] J. K. Hulm, Proc. Roy. Soc. (London) **A204**, 98 (1950). See L. Tewordt, Phys. Rev. **129**, 657 (1963), Fig. 3 for comparison of thermal conductivities of various materials.
[6] D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. **112**, 1888 (1958).

The preliminary reports of this investigation[7] indicated an anomalously large zero-temperature energy gap of approximately $8kT_c$ for lead. Further investigation revealed that this anomalous result was due to the presence, in the superconducting state, of an amplitude-dependent attenuation. This has also been reported on briefly.[8] Because there will certainly be questions regarding this effect, we describe crystal preparation, apparatus, and data analysis in some detail in Sec. II A, B, and C, respectively. The low-amplitude results and amplitude-dependent effects are discussed in Secs. III A and B and such conclusions as can be reached from this work are contained in Sec. IV.

II. EXPERIMENTAL

A. Crystal Preparation

The single-crystal lead ingot, from which the specimens used in this investigation were cut, was obtained from the Unimet Corporation. Table I shows the results of a spectrographic analysis performed on a small sample cut from the original single crystal. The ingot was set in litharge and glycerine paste, which hardened and supported it. Cubical specimens, approximately 0.5 in. on a side, were carefully cut from it using a jeweler's saw. Flat, parallel faces were then prepared on these specimens, again mounted in litharge and glycerine, by fly-cutting. In this process, the cutting tool passes the

TABLE I. Spectrographic analysis of lead specimens.[a]

Copper	0.0005%	Zinc	<0.0005%
Arsenic	<0.0005%	Antimony	<0.0005%
Bismuth	0.00008%	Iron	<0.0001%
Tin	<0.0005%	Silver	<0.00001%

[a] A semiquantitative analysis carried out by the Crobaugh Company, Cleveland, Ohio.

[7] R. E. Love and R. W. Shaw, Bull. Am. Phys. Soc. **8**, 420 (1963).
[8] R. E. Love and R. W. Shaw, Rev. Mod. Phys. **36**, 260 (1964).

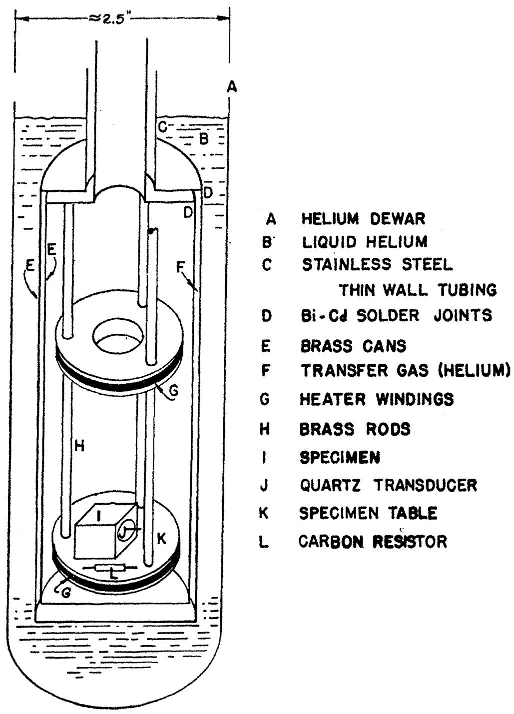

FIG. 1. Cutaway of specimen chamber.

specimen at high speed, always taking a very shallow cut. In this way the depth of the damaged region can be made very small. The specimens were then hand lapped with successively finer grinding compounds until the two surfaces were flat and parallel to within ±0.0001 in. and perpendicular to the desired crystallographic direction to within one degree. A light etch following the lapping was sufficient to produce good single-crystal x-ray pictures, indicating that most of the surface damage caused by the fly-cutting had been removed by the lapping. Finally, before placing the specimens in the cryostat, they were annealed in vacuum at 275° for about 8 h.

The data presented in this work were obtained from two specimens prepared as outlined above. On one, two surfaces were perpendicular to the [100] direction; on the other, two surfaces were perpendicular to the [111] direction and two were perpendicular to the [1$\bar{1}$0] direction. The transducers were 10- and 5-Mc/sec fundamental x-cut quartz of $\frac{1}{4}$ in. diameter obtained from the Valpey Crystal Corporation. They were chrome- and gold-plated on one side only and bonded to the specimens with Dow Corning 20 000 centistokes silicone fluid. The electrical connection to the transducer was made by either spring contact or a phosphor-bronze wire soldered to the transducer by the manufacturer.

B. Apparatus

The specimen chamber is shown in cutaway view in Fig. 1. It consisted of a double-walled brass can in which the pressure of helium gas between the walls and within the inner can could be varied. In general the pressure within the inner can was kept fairly high to aid in maintaining isothermal conditions within the can. The wall pressure, together with the heater power input, was varied to obtain temperatures between 1.3 and 20°K. Data at higher temperatures were taken during slow drifts at the beginning and end of the runs.

Temperature was measured in the range below 20°K by an Allen Bradley 56-Ω, $\frac{1}{2}$-W carbon resistor cemented to the brass specimen table adjacent to the specimen. The resistor was calibrated below 4.2°K against the vapor pressure of liquid helium[9] condensed in the inner can, at 7.185°K against the transition temperature of the lead,[10] and above 10°K against a Chromel-constantan thermocouple.[11] The empirical equations of Clement and Quinnel were used for interpolation.[12] Above 20°K the chromel-constantan thermocouple was used directly. An amplifier circuit automatically controlled the temperature by varying the current in the heater coils in response to the unbalance of a Wheatstone bridge in which the carbon resistor formed one of the arms. Temperature drifts were arranged by manual adjustment of heater input and wall vacuum. The specimen was held in place by upright brass plates cushioned from the specimen by a layer or two of tissue paper. The rf pulses were carried by any of three $\frac{1}{16}$-in.-diam thin-wall stainless-steel tubes spaced in the inner-can support tube. Multiple specimens and/or transducers were occasionally employed during the same run by making use of these multiple leads.

During measurements in the superconducting state the earth's magnetic field was cancelled to within approximately 0.03 G by external coils. Prior to such measurements the specimen was always taken above T_c in this small field in order to avoid trapped flux. For normal-state measurements, a 12-in. Harvey Wells magnet was rolled up to the cryostat. It provided fields uniform to within a few parts in 10^5 over the specimen volume.

A block diagram of the circuitry associated with the ultrasonic measurements is shown in Fig. 2. The pulsed oscillator, superheterodyne receiver, first detector and oscilloscope were contained in a single instrument, a Speery ultrasonic attenuation comparator. For purposes of investigating the amplitude dependence an additional attenuator was sometimes inserted just outside the pulsed oscillator. This allowed the input pulse amplitude to be varied without change in pulse shape or frequency. The techniques and pitfalls of this type of measurement have been discussed by Morse.[13]

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[9] Natl. Bur. Std. (U.S.) Monograph 10, (1958).
[10] W. B. Pearson and I. M. Templeton, Phys. Rev. **109**, 1094 (1958).
[11] A calibration of this type of thermocouple against a platinum resistor was kindly supplied by T. G. Nilan and A. V. Granato (private communication).
[12] J. B. Clement and E. H. Quinnell, Rev. Sci. Instr. **23**, 5 (1952).
[13] R. W. Morse, in *Progress in Cryogenics* (Heywood and Company, Ltd., London, 1959), Vol. I, p. 219.

The attenuator at the input provides an accurate and convenient means of measuring the relative echo amplitude. More will be said about this in Sec. II C. Both the diode limiter circuit and the tunable preamplifier (Arenberg PA-620-B) were designed to ensure that the receiver had recovered from the initial pulse before the first echo returned. The pulse widths were approximately 2 μsec and the round trip time about 10 μsec. The gate-and-detector circuit was similar to that described by Kamm and Bohm.[14] Its function was to allow only one particular echo to pass on into the second detector. The output of this unit drove the recorder pen.

C. Data Analysis

The data were obtained, for the most part, in the form of strip-chart recordings of the gated echo as the field or temperature was slowly varied. The amplitude of the record was translated into decibels by stopping the drift at least once on each record and inserting various attenuations by means of the calibrated attenuator. This dropped the entire echo train by known factors and furnished a "ruler" against which to measure the echo amplitude. If a_n and b_n are the amplitudes in dB of the echo at two different times during a record, the change in attenuation between these two times is given by

$$\alpha'_b - \alpha'_a = ((a_n - b_n)/nD)\ \mathrm{dB/cm}\,, \qquad (2)$$

where D is the round trip distance in cm and n is the echo number in the series. The expression assumes that the initial pulse is the same when a_n and b_n were measured. This was checked by monitoring the initial pulse during drifts and, more accurately, by recording a single echo under constant conditions. These indicated a constancy of the initial pulse to within 0.1% over times comparable to those required to obtain a single record.

In order to obtain absolute attenuation values[15] the nth echo was gated and the recorder deflection noted.

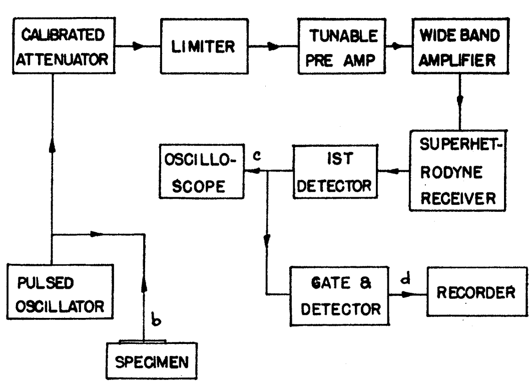

FIG. 2. Block diagram of circuitry for ultrasonic measurement.

[14] G. W. Kamm and M. V. Bohm, Rev. Sci. Instr. 33, 957 (1962).

[15] Morse (Ref. 13) has discussed the greater importance of relative attenuation values in measurements such as these.

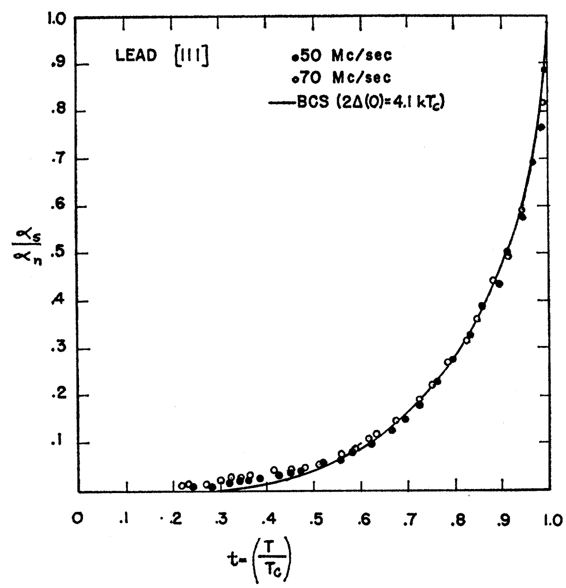

FIG. 3. Ratio of electronic attenuations versus reduced temperature for propagation in the [111] direction. The solid line illustrates the temperature dependence predicted by the BCS theory for the indicated zero-temperature energy gap.

An earlier echo, number j of the echo train was then gated and the attenuation required to bring this deflection down to that of the nth echo was noted. The absolute attenuation was then given by the inserted attenuation divided by the number $(n-j)$. An echo train was, of course, not acceptable unless the results of such a procedure were approximately the same for all n and j. Such was the case for the results to be reported in Sec. III A. By this technique, α'_a of Eq. (2) could be measured and α'_b determined from the record.

The primed attenuation of Eq. (2) is the total observed attenuation while Eq. (1) predicts the ratio of electronic attenuations. These electronic attenuations were determined by subtracting a background attenuation α_B from the experimentally determined quantities, i.e.,

$$\alpha_s/\alpha_n = (\alpha'_s - \alpha_B)/(\alpha'_n - \alpha_B)\,. \qquad (3)$$

α_B was determined by extrapolating to $T=0°$K the superconducting curves taken for the smallest possible initial pulses. At that point electronic attenuation is expected to be zero.

In addition it has been assumed that α_B is constant throughout the superconducting range. As the results are presented, the justification for this assumption will be pointed out. Below T_c, the normal-state attenuation α'_n was obtained as follows: First, a magnetic field of 900 G was applied parallel to the propagation direction and the attenuation measured as the temperature was slowly varied. Then the attenuation was measured as a function of magnetic field at several fixed temperatures. These latter curves were then extrapolated to zero applied field in order to provide a correction curve

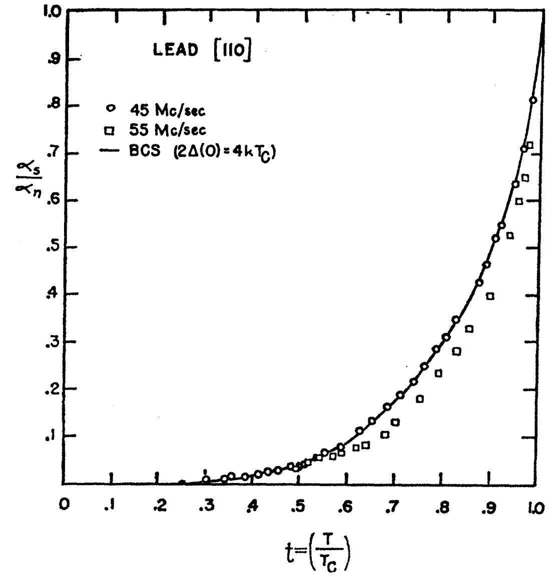

FIG. 4. Ratio of electronic attenuations versus reduced temperature for propagation in the [110] direction.

for the 900-G drift. Once α_s/α_n versus temperature was determined Eq. (1) was used to calculate $2\Delta(T)/kT_c$.

III. RESULTS AND DISCUSSION

A. Low Amplitudes

The most reliable low-amplitude results of this investigation are shown in Figs. 3 through 6. It is apparent that the fit to the BCS temperature dependence with a $2\Delta(0)$ of approximately $4kT_c$ is good for reduced temperatures ($t=T/T_c$) above 0.5. The exception to this is the 55-Mc/sec curve for the [110] propagation direction. The reason for this and for the deviations below $t=0.5$ will be discussed below. In comparing our results with the BCS and related theories, we shall confine our attention to the 45 Mc/sec, [110] and the 50 and 70 Mc/sec, [111] data for $t\geq 0.5$. We have not observed significant magnetoacoustic resonances in this temperature range. This, as well as the fact that α_n varies rapidly with temperature in the same range, indicates the ql (the product of the sound wave propagation vector with the electron mean free path) is less than one.[13] Under such circumstances, the attenuation cannot be attributed to a particular group of electrons and we must look upon these energy gap results as representing an average over the entire Fermi surface. Table II summarizes the best estimates of the zero-temperature energy gap for propagation in each of the three major crystallographic directions in lead. These values favor the strong coupling theory of Thouless[16] which predicts $2\Delta(0)=4kT_c$, over the BCS theory. However, as can be seen in Fig. 6 the normalized temperature dependences of the theories are quite similar and the present measurements do not lend support to one over the other.

[16] D. J. Thouless, Phys. Rev. 117, 1256 (1960).

TABLE II. Summary of zero-temperature energy gap for three directions of longitudinal sound-wave propagation in lead.

Direction	Frequency (Mc/sec)	$2\Delta(0)/kT_c$
[111]	50	4.1±0.1
	70	4.1±0.1
[110]	45	4.0±0.1
	55	<4.8[a]
[100]	50	<5.1[a]
	70	<5.1[a]
Best estimate[b]		4.1±0.2

[a] The 55-Mc/sec [110] data shown in Fig. 4 are an example of how these values were obtained. In such cases, the voltage applied to the transducer is probably not sufficiently small to eliminate the error caused by the amplitude-dependent effect. See text for more detail.
[b] The $\pm(0.1)kT_c$ error refers to the internal consistency of the data, whereas the $\pm(0.2)kT_c$ error on the best estimate includes the possibility of systematic errors.

The scatter in the data at low temperatures is due to instrumentation noise and is significant only in that it limits the accuracy with which the zero-temperature energy gap might be determined from the low-temperature data. The systematic deviations are of more concern, however. They may be due to (a) the assumption of a constant background, (b) the error in α_n arising from the difficulty of determining $\alpha(H=0)$ from the $\alpha(H)$ versus H curves, or (c) the fact that the attenuation in the superconducting state is amplitude-dependent. This last possibility will be discussed in Sec. III B.

First, we consider the assumption of a constant background. Actually, it is difficult in the present case to rigorously justify this assumption by direct experimental observation, i.e., the attenuation above T_c is always greater than α_B and α_n is not constant below T_c. However, the good agreement with the BCS theory

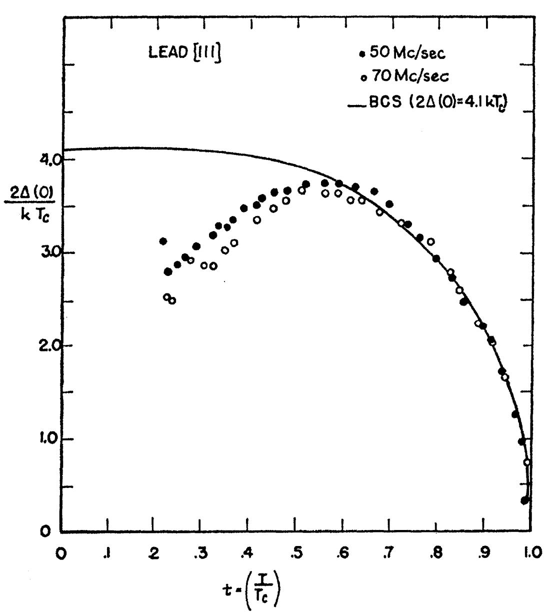

FIG. 5. Energy gap in units of kT_c derived from data of Fig. 3.

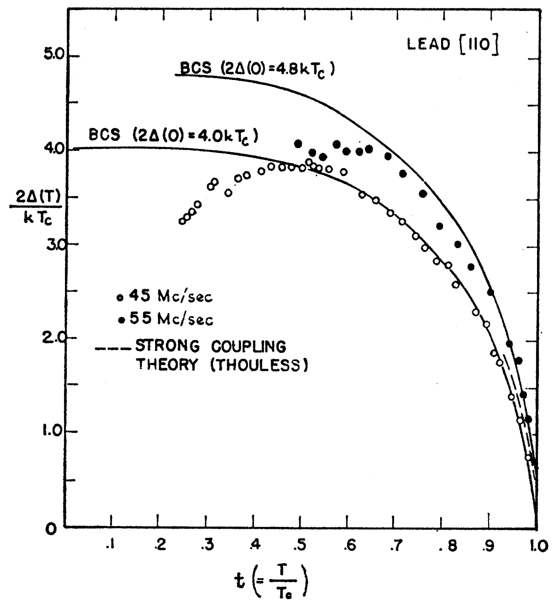

FIG. 6. Energy gap in units of kT_c derived from data of Fig. 4.

just below T_c, in those cases where we believe the amplitude-dependent effect to be small, gives some support to this assumption. Also, although the attenuation above T_c never drops to α_B, it does decrease monotonically with increasing temperature up to a temperature of about 70°K. At this point, the attenuation is only 1 or 2 dB/cm above α_B. Since most attenuation mechanisms, other than electron damping, result in an attenuation which increases with increasing temperature, it is reasonable to assume that they give rise to variations which are small below 70°K and negligible below T_c. Thus all observed changes will be considered electronic.

Next we consider α_n. Several investigators have carried out magnetoacoustic calculations,[17–19] all of which predict that the attenuation should be independent of H in the case of longitudinal waves with H parallel to the propagation direction. This is in serious disagreement with our results and those of Mackintosh for lead[20] and hence no attempt was made to employ the results of such calculations to determine $\alpha_n(H=0)$. Instead the extrapolation was based upon the systematic variation the $\alpha(H)$ versus H curves exhibited as a function of temperature. Above 5°K, almost no correction to the temperature drifts taken at 900 G was needed, since the attenuation remained relatively constant up to 9 kG. At lower temperature our results resembled those of Ref. 20 and the correction became more difficult to determine. However, since the normal-state attenuation was quite large at these temperatures (10 dB/cm or more), missing $\alpha_n(H=0)$ by as much as $\frac{1}{2}$ dB/cm leads to corresponding error in α_s/α_n of only 5% or less. A fairly good approximation in the same range, based upon $\alpha_s \ll \alpha_n$, yields an error of the same percentage in Δ. The deviations at low temperatures on Figs. 5 and 6 are closer to 25% and it is highly unlikely that they reflect errors in $\alpha_n(H=0)$.

B. Amplitude-Dependent Effects

We now come to the third possible explanation for the deviations of our data from the BCS temperature dependence. This explanation involves the dependence of the attenuation upon the amplitude of the ultrasonic pulse which has been observed in the present experiments. We first present the characteristics of this dependence and then show how it can lead to erroneous values of the energy gap.

The effect is most directly illustrated by the series of detected echoes seen on the oscilloscope. Figure 7 shows three such echo trains traced for clarity from photographs. These were taken with the indicated voltages at the transducer, adjusted by means of an attenuator at the output of the pulsed oscillator. The attenuator at the receiver was then adjusted until the height of the first echo was 6 cm on the scope face. Thus, the receiver saw the same magnitude of echo voltage in each case. In addition, since the pulsed oscillator and receiver controls were not adjusted the pulse shape and frequency remained constant. The echo trains clearly show a faster decay (indicating a higher attenuation) when the amplitude of the pulse in the specimen is larger. More detailed studies have led to the conculsion that the attenuation is a function of the instantaneous amplitude in the sample, i.e., if the oscillator attenuator

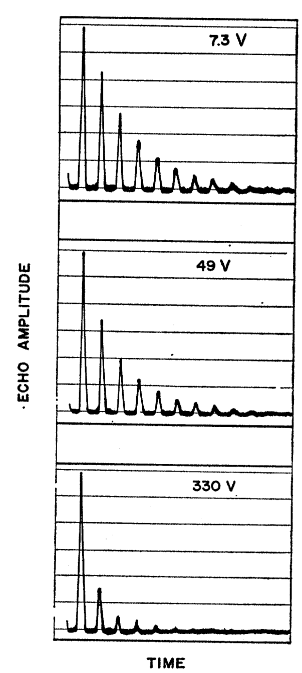

FIG. 7. Traces of oscilloscope photographs showing the detected echos as a function of time for the indicated peak-to-peak initial-pulse voltages at the transducer. A 30-Mc longitudinal pulse was being propagated in the [100] direction at 2.69°K.

[17] M. H. Cohen, M. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960).
[18] A. B. Pippard, Proc. Roy. Soc. (London) **A257**, 165 (1960).
[19] S. Rodriguez, Phys. Rev. **112**, 80 (1958).
[20] A. R. Mackintosh, Phys. Rev. **131**, 2420 (1963).

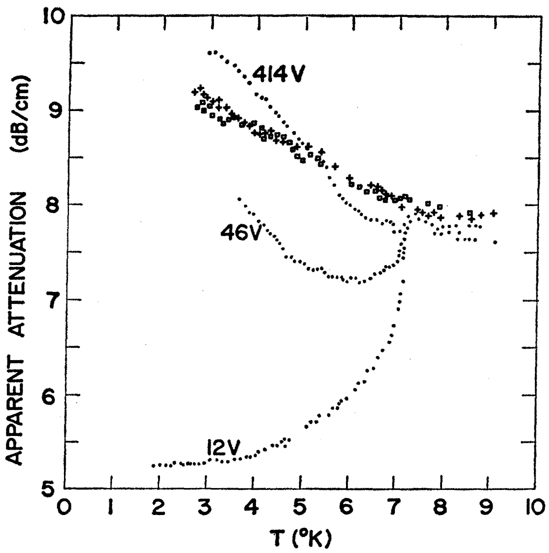

FIG. 8. Apparent attenuation (see text for discussion) versus temperature for various initial-pulse amplitudes. The 30-Mc/sec longitudinal pulse was propagated in the [111] direction. The curves labeled by transducer voltages are all in the superconducting state while the crosses (+) and squares (□) correspond to normal-state data with 12 and 414 V on the transducer, respectively. Some points near T_c have been omitted.

only is changed the attenuation at comparable echo amplitudes will be the same.

The results of analyzing data in the usual way (see Sec. II C) are shown in Fig. 5 of Ref. 8 for 10 Mc/sec in the [111] direction and in Fig. 8 of the present paper for 30 Mc/sec in the same direction. The ordinates of these figures are not attenuation coefficients in the usual sense because, in the presence of an amplitude-dependent absorption, the attenuation varies along the echo train. The ordinates may be thought of as an average attenuation between the initial pulse and the gated echo, or simply as the relative amplitude, in dB, of the gated echo. The important points to notice in Fig. 8 are the dependence of the apparent attenuation upon pulse amplitude in the superconducting state and the total absence of such an effect in the normal state. This normal-state behavior has been a general rule in our observations. Note also that the apparent relative attenuation in the superconducting state has been observed to rise higher than the normal-state attenuation.

For purposes of determining an energy gap, we used the lowest possible transducer excitation voltage (<10 V) consistent with obtaining a useful signal. In all cases, we found that the agreement with BCS improved as the excitation voltage applied to the transducer was decreased. The amplitude dependence exhibited the following empirical behavior:

(1) The apparent attenuation was always larger than or equal to the actual attenuation, i.e., the attenuation at minimum amplitude.

(2) Down to the lowest temperatures attainable with our apparatus, about 1.3°K, and for relatively low excitation voltages, the apparent attenuation deviates more strongly from the actual attenuation as the temperature is lowered.

These two properties are important in understanding qualitatively the effects of amplitude dependence upon energy-gap determination.

We first consider the consequences of a small amplitude-dependent effect on the α_s/α_n versus t and $2\Delta(T)/kT_c$ versus t plots. In this case, the experimentally measured attenuation may not deviate from that predicted by the BCS theory until quite low temperatures are reached. This deviation is difficult to detect on the usual α_s/α_n versus t plots because the scatter in the data becomes comparable to the changes expected in α_s. However, in such cases, the presence of an amplitude-dependent effect is clearly indicated on the energy gap plots. See, for example, the 45-Mc/sec data in Figs. 4 and 6. This is in accord with the second point in the previous paragraph, i.e., a small amplitude-dependent effect produces, at low temperatures, an apparent relative attenuation which is slightly greater than the attenuation BCS would predict. This leads to a corresponding falloff of $2\Delta(T)/kT_c$. In Fig. 9, the 45-Mc/sec [110] data have been plotted on an expanded scale to illustrate this effect. Using BCS theory and experimental values of α_n', the α_s' required to make the energy-gap versus reduced-temperature plot on Fig. 6 fit the BCS theory for a zero-temperature energy gap of $2\Delta(0)=4kT_c$ has been calculated. The purpose of comparing theory and

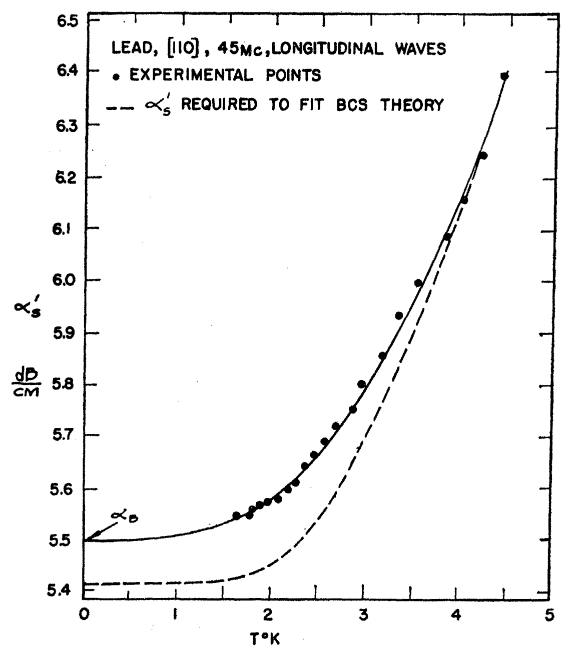

FIG. 9. Experimental α'_s curve versus temperature. The dashed line was computed using BCS theory with a zero-temperature energy gap of $4kT_c$ and experimental α_n values.

experiment in this way is to show that: for low amplitudes the amplitude-dependent effect produces an additional apparent attenuation at low temperatures, resulting in an error in the *shape* of the α'_s curve which is far more significant than the corresponding error generated in α_B.

Now let us consider the consequences of a somewhat larger amplitude-dependent effect. In this case, in addition to an error in the shape of the α'_s curve, the apparent background attenuation becomes significantly too large. This shows up immediately in the α_s/α_n plots as a too rapid drop near T_c. The 55-Mc/sec data in Fig. 4 are an example of this effect. Notice here that, due to the subtraction from all attenuation values of a background attenuation which is too large, the amplitude dependent effect causes an apparent error at high values of reduced temperature, i.e., there would probably be no error in this temperature range if the true background attenuation were used to compute α_s/α_n.

The presence of the amplitude dependence in the superconducting state is therefore able to account qualitatively for the deviations from the BCS theory present in Figs. 3 through 6. We conclude that this is the source of the deviations.

It is of interest to compare our data on lead with similar measurements made on other elements. Ultrasonic measurements by other workers on lead, mercury, tin, and indium exhibit deviations from the BCS theory similar to those associated with amplitude dependence in the present work. Bömmel's data on lead[1] are very similar to some of the data obtained earlier in this investigation.[7] At that time, we reported an energy gap of approximately $8kT_c$. Since then we have found, in cases such as reported by Bömmel (where the α'_s curve is flat between 5 and 3.5°K), that, on carrying the measurements to lower temperatures, the α'_s begins to rise again, indicating the presence of a strong amplitude dependent effect. The results of Chopra and Hutchison[21] on mercury, those of Morse *et al.*[22] on tin and indium, and those of Saunders and Lawson[23] on thallium, show deviations from BCS theory which are similar to those observed here at low amplitudes.

At the present time, we are not sure of the cause of the amplitude-dependent effect. The experimental observations in lead which a theory describing this effect must explain are as follows:

(1) All evidence to date indicates that the effect is present only in the superconducting state. (2) In the cases investigated to date, the effect is present for both shear and longitudinal waves. (3) We have made measurements at 10, 15, 25, 30, 35, 45, 50, 55, 65, and 70 Mc/sec and in all cases the effect was observed to be present. (4) We have observed the effect in the [100], [110], and [111] crystallographic directions. (5) To within 0.5%, the acoustic velocity does not depend upon amplitude. (6) We have not observed second or third harmonic generation from a 15 Mc/pulse. We estimate that the second harmonic is at least 40 dB down from the fundamental.

The fact that this effect has not been reported previously is rather puzzling. It is possible, however, that the effect may be small except in superconductors such as lead where the electron-phonon interaction responsible for superconductivity is thought to be particularly strong. Strong coupling implies a strong interaction between the electron system and the lattice, hence a small lattice perturbation due to the passage of an acoustic wave may have a large effect on the effective electron-electron interaction. It is difficult, however, to see how such a mechanism could give rise to an attenuation in the superconducting state which is greater than the corresponding attenuation in the normal state.

At the frequencies used in this investigation, dislocation damping is another possible absorption mechanism. It is tempting to invoke a dislocation mechanism to explain our observations for two reasons. First, it is well known that dislocations are responsible for amplitude dependence at higher temperatures and, second, lead is quite soft compared to other elemental superconductors, implying the presence of a large number of dislocations. However, near T_c, where the attenuation is not too different from that in the normal state, it is difficult to see how dislocations could give rise to a very large amplitude effect in the superconducting state, while, at the same temperature, in the normal state there is no measurable amplitude dependence. Recent preliminary measurements by Deaton,[24] indicating no amplitude dependence in a spark cut lead sample, would seem to lend weight to the dislocation source idea, since his sample should contain far fewer dislocations than ours. Tittmann and Bömmel[25] have recently confirmed the existence of an amplitude-dependent attenuation in lead. They propose a dislocation mechanism.

CONCLUSIONS

The results of this investigation indicate that the average value of the zero-temperature energy gap in lead is $2\Delta(0)=(4.1\pm0.2)kT_c$, in substantial agreement with measurements by other techniques. In addition, the temperature dependence of the gap is in good agreement with the BCS theory for reduced temperature above 0.5 if the above value of $2\Delta(0)$ is used. Due to deviations of the data from this temperature dependence at lower temperatures and to frequency limitations of the present apparatus no measure of anisotropy was obtained.

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These deviations are limited to $T/T_c<0.5$ only by working at low amplitudes. For higher ultrasonic amplitudes, they spread throughout the superconducting range. The amplitude-dependent attenuation which gives rise to the deviations appears to be a property of the superconducting state only. This amplitude dependence, while certainly not as large in the other superconductors thus far studied by ultrasonic attenuation may still be present in them and give rise to some of the other observed deviations from the theoretical temperature dependence.

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Temperature and Field Dependence of Hyperfine Fields and Magnetization in a Dilute Random Substitutional Ferromagnetic Alloy: $Fe_{2.65}Pd_{97.35}$†

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The assumption of proportionality between hyperfine magnetic field and bulk magnetization, which has been shown to obtain in elemental ferromagnetic Fe, is shown to apply as well in the random substitutional cubic dilute ferromagnetic alloy $Fe_{2.65}Pd_{97.35}$. The same sample of this alloy was studied by means of both the Mössbauer effect and bulk magnetization. The Mössbauer data represent the first detailed study of the hyperfine field in a ferromagnet in the presence of large external fields. Proportionality between the bulk magnetization and the hyperfine field is found over an extended range of temperature and applied external magnetic field. Extrapolations of the bulk-magnetization data to yield the Curie point disclose significant discrepancies with the Mössbauer results when standard extrapolation techniques are used. Good agreement is obtained if one assumes that the magnetization varies near the Curie temperature θ as $(\theta-T)^\beta$ with β between $\frac{1}{4}$ and $\frac{1}{3}$. This type of behavior suggests that the sample may be describable in the region of the Curie temperature by recent Padé-approximant calculations based upon Ising and Heisenberg models. The temperature variation of the hyperfine field is compared to molecular-field calculations in both zero and nonzero external fields. Although qualitative agreement is found, there are significant quantitative discrepancies, indicating the inapplicability of the molecular-field model. A description is included of the Brookhaven cryogenic Mössbauer apparatus involving a variable-temperature cell mounted inside a high-field superconducting magnet.

I. INTRODUCTION

The temperature dependence of the hyperfine magnetic field in ferromagnetic materials has been studied by means of the Mössbauer technique in pure Fe and at Fe in $Fe_{08}Pd_{92}$ by Nagle *et al.*[1] and by Preston *et al.*[2] in pure Fe. These measurements were performed in zero external magnetic field. Comparison between the Mössbauer measurements and bulk-magnetization measurements showed that the two quantities vary with temperature in the same way. That is, the hyperfine field is proportional to the bulk magnetization. This result indicates that for fixed sample composition the magnetic hyperfine interaction is temperature-independent and that in addition the spatial distribution of the magnetic moment is temperature-independent.

We have extended the study of Nagle *et al.*[1] by applying external magnetic fields to a relatively dilute ferromagnetic Fe-Pd alloy, $Fe_{2.65}Pd_{97.35}$, and have used the same sample for both Mössbauer and bulk-magnetization studies We find excellent agreement between the temperature dependence of these two quantities over a wide range of temperature and externa magnetic field. The region close to the Curie temperature (about 90°K) and in low external fields is of especial interest. In this region the sample magnetization and the hyperfine field appear to vary with tem-

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