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ULTRASONIC ATTENUATION MEASUREMENTS
IN SUPERCONDUCTING METALS

by

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INTRODUCTION

The purpose of the investigations reported in this thesis was to study the attenuation of high frequency sound pulses in metallic tin and tantalum in the very low temperature region near their super conducting transitions and to study the temperature dependence of sound velocity and the elastic moduli of metallic beryllium. Laboratory apparatus for producing sound pulses of frequencies in the range of 9 to 11Mc/sec. and duration from 2 to 20 microseconds was constructed for these studies. The recent development of electrical circuits for producing very short radio frequency pulses has provided the knowledge needed in the design and construction of this apparatus.

If an electro-mechanical transducer such as a piezoelectric quartz crystal is excited at its resonant frequency by a short duration radio frequency electrical pulse, the sound pulse generated will be propagated into the medium in which the transducer is imbedded. Alternately when a sound wave traveling through the medium strikes the transducer, it results in the generation of an electrical signal. Thus such a transducer may be used as both the generator and detector of sound waves. The combination of this principle, together with the necessary electrical generating and measuring equipment
is known as the ultrasonic pulse technique.

Using this technique it is possible to generate sound pulses of durations from less than one microsecond to many microseconds and frequencies from a few kilocycles per second up into the neighborhood of 100 megacycles per second.

In the laboratory application of the ultrasonic pulse technique, the electro-mechanical transducer is usually a flat disk type quartz crystal, either X-cut or Y-cut, which propagates respectively a compressional or shear sound wave in the direction perpendicular to the disk faces.

In order to measure the compressional or transverse velocity in a solid specimen the specimen must have two parallel faces. The quartz is cemented to one face while the other face serves as a reflector. Twice the perpendicular distance between the faces is the round trip distance traveled by the sound wave. In liquids and gases a metal reflecting surface is placed perpendicular to the path of the sound beam.

The electrical equipment consists of a special cathode ray oscilloscope which can be used to time the arrival of the sound pulses that have made the round trip through the specimen. The velocity is determined from the measured times of arrival of the pulses and the round trip distance traveled. Certain materials, e.g. simple liquids like water and the light metals such as beryllium
propagate sound pulses of a few microseconds so well that it is possible to obtain velocity measurements accurate to one part in ten thousand. However in the polycrystalline heavy metals the scattering and absorption of the sound by the crystallites in many cases, distort the shape of the sound pulse so that velocity measurements are only accurate to a few percent.

A RESUME OF ULTRASONIC TECHNIQUES

Prior to the development of electrical circuits for pulsing, the two principle ultrasonic devices were the composite oscillator, essentially that developed by Balamuth for velocity measurements in solids and the acoustic interferometer. The composite oscillator employs a piezoelectric quartz rod, onto which electrodes are plated, cemented to a rod of the solid specimen. This assembly is then supported by fibers attached to the quartz at its vibrational nodes. The quartz, excited near its resonant frequency by continuous waves from an R.F. oscillator, sets up standing waves in the composite oscillator. When the frequency of the driving oscillator is equal to the one of the resonant frequencies of the composite oscillator, the reactance of the quartz crystal will increase to a maximum and the current in the radio frequency output circuit will decrease to a minimum. Several
frequency shifts permits a determination of the velocity through the formulae in Appendix I, accurate to a few per cent.

Using Balamuth's composite oscillator principle, Rose$^2$ obtained the elastic constants of sodium chloride single crystals from $80^\circ K$ to $300^\circ K$. Later Hunter and Siegel$^3$ employed the composite oscillator to measure the elastic constants of sodium chloride single crystals in the high temperature range from $300^\circ K$ to $800^\circ K$.

The acoustic interferometer is a well known device for measuring ultrasonic wavelengths in liquids and gases. It uses a metal reflecting plane placed opposite the vibrating quartz with the gas or liquid in between. Standing waves are set up in the medium. Moving the reflector changes the standing wave condition at the quartz crystal through loops and nodes, resulting in changes in the R.F. current to the oscillator. Hence integral multiples of half wavelengths can be measured and the velocity determined. This technique is widely used for measurements in liquids and gases.

Recently van Itterbeek$^4$, using the acoustic interferometer, measured the velocity in liquid hydrogen as a function of the percent parahydrogen to normal hydrogen. He found that the transformation from normal hydrogen to 100% parahydrogen was very strongly catalyzed by the ultrasonics, if a small amount of oxygen was added
to the hydrogen. This indicates a coupling of the spin system of the $\text{H}_2$ molecule with either vibrational or rotational modes of the molecule, at least in the presence of the oxygen. He observed a velocity increase as the percent parahydrogen increased.

The ultrasonic pulse technique may be used in several ways. Pellam and Squire\textsuperscript{5} used the pulse echo method to measure velocity and absorption of 15Mc/sec. sound pulses in liquid helium from 4.2$^0\text{K}$ to 1.5$^0\text{K}$. They employed a movable reflector operated by a screw in order to conveniently set the round trip distance traveled by the sound wave. This movable reflector proved to be an accurate and convenient aid in the measurement of the sound absorption since moving the reflector a distance necessary to reduce the pulse amplitude a measured amount immediately gives the attenuation coefficient.

In the pulse-echo technique, discussed in detail on page 14 for measurement in solids it is necessary to have at least two pulses of sufficient amplitude to be observable in order to reliably measure the attenuation of the sound. Quite often this is a difficult experimental condition to satisfy and depends strongly on the structure characteristics of the solid. In general it is very difficult to obtain good results in the heavier metals having polycrystalline structure.

Roth\textsuperscript{6} used the pulse-echo technique to measure
the velocity and absorption as a function of frequency and grain size in polycrystalline specimens of magnesium and aluminum. His experimental attenuation measurements were quite accurate enough to clearly obtain the dependence of attenuation with frequency.

Hughes developed a technique for measuring both the bulk and the shear moduli of metal rods, utilizing an effect observed by Mason and McSkimin. A plane ultrasonic compressional wave pulse is sent into the rod by a quartz crystal at one end. The wave travels through the rod and the pulse is detected at the other end by another quartz crystal. Part of the compressional wave front strikes the cylindrical walls of the rod at grazing incidence. This part of the wave is converted into a transverse wave which travels across the rod at an angle $\Theta$ with the normal to the cylindrical surface where

$$\sin \Theta = \frac{\text{transverse wave velocity}}{\text{compressional wave velocity}}$$

On striking the opposite wall the wave is converted back into a compressional wave where it either travels on to the receiving crystal resulting in a delayed pulse or undergoes additional surface reflections resulting in additional pulses to the crystal that are delayed even further in time with respect to the initial compressional pulse. Measurement of the initial and delayed times of
arrival of the pulses in rods of different lengths and
diameters allows calculation of the elastic moduli. The
formulae are given in Appendix I.

The values of the elastic constants of solids
are of theoretical significance. It is possible to
obtain the value of the microscopic elastic constants
needed in the theoretical calculation of the vibrational
spectra of a solid from the experimentally measured
values of the elastic constants.

**MEASUREMENTS ILLUSTRATING THE ULTRASONIC TECHNIQUE**

**IN THIS INVESTIGATION**

The present research is concerned mainly with
measurement of the velocities of longitudinal and
transverse ultrasonic waves in solids at low temperature
so that the adiabatic elastic constant and or moduli can
be determined and with the measurement of the attenuation
of the high frequency sound in metals at low temperature.
However, the technique can be readily adapted to
measurements of velocity and attenuation in liquids.

As an illustration of the accuracy of velocity
measurements, Figure 1-a shows a photograph of the
oscilloscope time base trace for sound pulses traveling
in a narrow glass tube filled with water. The round trip
distance traveled by the pulses was 28.60 cm. The total
time interval from the left to the right edge of the trace
is 1000 microseconds. The first echo is shown in Figure 1-b on an expanded scale of 25 microseconds duration. The time delay, after the start of the oscilloscope trace shown the left edge of Figure 1-a, to the vertical slope on the leading edge of the pulse on the expanded scale was measured as 192.5 microseconds. The leading edge of the second echo was at 385.0 microseconds.

The velocity of 10Mc/sec. sound pulses in the water tube comes out to be 1.47 \times 10^5 \text{cm/sec}. The principle error here was in the measurement of the distance.

Figure 1-c shows a multiple echo oscilloscope trace at room temperature in a metallic beryllium cylinder 15.209 cm in length and 1.5 inches in diameter. The time interval between echos, averaged over the 8 echos shown on the 100 microsecond sweep was 12.10 \pm 0.01 microseconds. This gives a velocity of 1.254 \pm 0.001 \times 10^6 \text{cm/sec.} or an accuracy of better than one part in a thousand. The small signals appearing after each echo are reflections from the cylinder walls which increase in amplitude toward the right side of the trace.

In Figure 1-a the exponential decay of the amplitude of pulses in the tube of water is clearly indicated. Electrical attenuation inserted in the line to the receiver reduces the height of the echos. The number of decibels attenuation required to reduce the first echo to the level of the second echo when no
Electrical attenuation is inserted gives the attenuation in decibels per 28.6 cm. The measured value for one cm was 0.45 dB.

ULTRASONIC ATTENUATION IN SOLIDS

There are many mechanisms contributing to the absorption of sound in solids. It is well known that polycrystalline solids attenuate sound more than single crystals due to the scattering of the sound wave by the grains making up the polycrystalline structure. Other mechanisms are thermoelastic relaxation and internal friction, plastic flow, heat conduction and magnetic effects in ferromagnetic materials. Thermoelastic relaxation is not important above about 100 kc/sec. and plastic flow and absorption due to magnetic effects are not of prime interest at the present. The factors of importance in the present research are the attenuations due to scattering and thermal conduction.

Energy losses due to scattering from grains are of course not encountered in single crystals. The attenuation due to scattering is important above about 1 Mc/sec. which puts it in the frequency range of pulsed ultrasonics.

The effect of scattering has been worked out by Mason and McSkimin who started out with Raleigh's
formula for the scattering of sound from a sphere:

\[ I_s = I_i \frac{\pi D}{R \lambda^2} \left[ \frac{\Delta k}{K} - \cos \theta \frac{\Delta \rho}{\rho} \right] \]

where \( I_s \) is the scattered sound amplitude, \( I_i \) is the incident amplitude in the plane wave, \( D \) is the particle volume, \( \lambda \) the sound wavelength, \( k \) the elasticity of the medium and \( \Delta k \) the difference in the elasticity of the scatterer and the medium. For scattering from grains the density difference \( \Delta \rho \) is neglected. The angle \( \theta \) is the angle measured from the direction of the sound beam at the particle to the observer. The term \( \frac{\Delta k}{K} \) is not zero because the crystallites present different stiffness coefficients depending on their orientations relative to the direction of the sound wave. The scattered energy is proportional to the square of the amplitude so that for a spherically symmetrical scattered wave the total scattered energy is

\[ E_s = I_i^2 \frac{\pi^2 D^2}{R^2 \lambda^4} \left[ \frac{\Delta k}{K} \right]^2 \iint R^2 \sin \theta d\theta d\phi \]

\[ = I_i^2 \frac{4\pi^3 D^2}{\lambda^4} \left[ \frac{\Delta k}{K} \right]^2 \]

If there are \( N \) randomly oriented grains in a volume element of thickness \( dx \) and effective cross section \( A \) to the sound beam direction, then the total
scattered energy is given by

\[ E_s' = I_i^2 \frac{4\pi^3}{\lambda^4} \sum_{j=1}^{N} D_j^2 \sum_{j=1}^{N} \frac{1}{N} \left[ \frac{\Delta K}{K} \right]_j^2 \]

if the grains are randomly oriented with respect to elasticity and \( D_j \) is independent of \( \left[ \frac{\Delta K}{K} \right]_j \). If \( \bar{D} \) is the average particle volume then

\[ \sum_{j=1}^{N} D_j^2 \approx N \bar{D} = \bar{D} A dx \]

Since \( \Delta I_i^2 \) is the incident energy in the sound wave

\[ E_s' = E_i \frac{4\pi^3 \bar{D}}{\lambda^4} \left[ \left( \frac{\Delta K}{K} \right)_{av.} \right] dx \]

The energy attenuation is of the form

\[ E(x) = E_0 e^{-\alpha x} \]

where

\[ \alpha = \frac{4\pi^3 \bar{D}}{\lambda^4} \left[ \left( \frac{\Delta K}{K} \right)_{av.} \right] \]

so that the attenuation coefficient for the amplitude of pressure wave is given by

\[ \beta = \frac{2\pi^3 \bar{D}}{\lambda^4} \left[ \left( \frac{\Delta K}{K} \right)_{av.} \right] \]

Mason and McSkimin have obtained a theoretical approximation to the inhomogeneity factor \( \sum_{j=1}^{N} \left( \frac{\Delta K}{K} \right)^2 = \left[ \left( \frac{\Delta K}{K} \right)_{av.} \right] \) by evaluating the average value of the elastic constants over the various orientations. Their experimental work on aluminum rods showed fair agreement for \( \bar{D} \) (about 0.9\( \lambda \)) but poor agreement for \( \bar{D} \) (about 0.5\( \lambda \)). On the other
hand Roth found a \( \frac{1}{D} \) dependence in polycrystalline aluminum and magnesium.

If the grain sizes are of the order of a wavelength then reflection of a portion of the sound wave may result. For a volume distribution of grains much larger than the average, multiple reflections may result in such a way as to return a complex wave front to the quartz crystal at the same instant the normal pulse reflected from the end face arrives. This results in bad pulse distortion and can cause inaccuracies in velocity measurements by obscuring the leading edge of the echo or completely obscuring the echo. In the present research, measurements in tantalum tend to indicate these effects. The echoes were so distorted that their leading edges could not be accurately located. In addition some echoes appeared at the wrong place on the oscilloscope indicating actual reflection from the inhomogeneities.

The mechanism of energy loss due to thermal conduction of heat out of the compressed portions of the sound wave is generally assumed to be described by the same equation as for gases (see Kittel for example). In terms of the diminution due only to thermal conduction, of pressure in a plane sound wave, the attenuation coefficient is

\[
\alpha_{\text{thermal}} = \frac{2\pi^2(\gamma-1)Kf^2}{\rho V^3 C_p}
\]
where $\gamma = C_p/C_v$, $K$ is the thermal conductivity, $f$ the frequency, $\rho$ the density, $v$ is the sound velocity and $C_p$ is the specific heat.

For solids in the liquid helium temperature range, $C_p = C_v$ so that $\gamma - 1$ is very nearly equal to zero. Thus at $T = 0^\circ K$, $\alpha_{th} = 0$ if this expression is correct. For $\gamma - 1$ very small, it should be possible to measure $\alpha_{th}$ by resorting to sufficiently high frequencies. If a single crystal is used then the principle mechanism for absorption will be that of thermal conduction.

For superconducting metals one should observe a discontinuity in $\alpha_{th}$ due to the known discontinuous increase in $C_p$ at the superconducting transition. Figure 2-a is a plot of the right hand side of the equation

$$\frac{\alpha_{th}}{(\gamma - 1)} = \frac{2\pi^2 K f^2}{\rho \sqrt{3} C_p}$$

for tin in the neighborhood of the superconducting transition. The experimental values of the thermal conductivity $K$ are taken from Radmaker's data in Figure 2-b. The measurements of $C_p$ for tin near the superconducting transition by Keesom and Kok are in Figure 2-c. The velocity was measured with the Rice Institute Low Temperature Laboratory ultrasonic equipment at a frequency of $10 M\alpha$/sec. and found to be $3.32 \times 10^5$ cm/sec.

The present research has been mainly directed
Fig. 2-a. Thermal attenuation in Tin at 10 Mc/sec
\( \alpha_t = \frac{2\pi^2 kT^2}{\rho \sqrt{\phi} \alpha_s} \) vs. Temp

Fig. 2-b. Thermal Conductivity of Tin (Bridgman)

Fig. 2-c. Atomic Heat of Tin (Keesom and Kok)
toward a measurement of the change in the attenuation coefficient in superconducting tin and tantalum. If the accuracy of attenuation measurements can be sufficiently improved then it should be possible, by such measurements, to determine the ratio \( \gamma = \frac{C_p}{C_v} = \frac{K_T}{K_S} \) where \( K_T \) and \( K_S \) are respectively the adiabatic and isothermal compressibilities. It is certainly not possible, with present techniques of measurement to determine \( \gamma \) by independent measurements of \( K_T \) and \( K_S \) in the very low temperature range because of the limits of accuracy of the existing methods.

**EXPERIMENTAL EQUIPMENT**

The operating principle of the ultrasonic pulse method is best described by reference to Figure 3. A crystal controlled timing circuit in the Du Mont oscilloscope starts the oscilloscope time base sweep and simultaneously sends a trigger pulse to the R.F. Pulsed Oscillator. The radio frequency pulse formed by the oscillator is amplified and passes through the impedance matching network to the quartz transducer, which introduces a pulsed sound wave into the solid. This sound wave is propagated through the solid to the bottom face where it is reflected and then returns to the
transducer forming a delayed electrical pulse. Since the radio frequency generating circuit is off when the echo signal arrives, very little leakage less occurs and most of the energy travels through the attenuator to the receiver.

The complete experimental apparatus is shown in Figure 3-b. The nomenclature and purpose is as follows.

A - Pulse Former, R.F. pulsed oscillator and amplifier

B - Broad Band Receiver - (Band width approximately 2Mc/sec. about 10Mc/sec. center frequency) and regulated power supply

C - \( \int \) - Network Strip Attenuator

D - Regulated Power and Bias Supply

E - Du Mont A/R-256-D Cathode Ray Oscillograph

F - Impedance Matching Network (Variable Inductance with 72 ohm coil input and variable capacitance)

G - Pumping Line to Mechanical Pump

H - Top Plate, Remote Control Shaft for lowering specimen onto electrical contacts

I - Inner Dewar Flask (Surrounding apparatus and receiving vessel for the liquid Helium)

J - Outer Dewar Flask - Contains liquid air for precooling as well as protecting liquid helium in the Inner Dewar Flask from thermal radiation.

K - Mercury Manometer for vapor pressure measurement of
the temperature.

L - Potentiometer for measurement of E.M.F. of thermocouple thermometers

The strip attenuator is a $\Pi$-network type made of pure carbon commercial resistors, which remain approximately purely resistive up to about 30M$\Omega$/sec. filed to the correct resistance and preserved with Glyptal varnish. It consists of 20, 20, 20, 10, 5, 3, 2, and 1 decibel $\Pi$-sections that can be switched in or out by double pole double throw switch. In the sketch $R = \frac{\alpha}{2 \times 0.4343} \sinh \left( \frac{\alpha}{2 \times 0.4343} \right)$

$R_1 = \frac{\alpha}{4 \times 0.4343} \coth \left( \frac{\alpha}{4 \times 0.4343} \right)$

$\alpha = \text{attenuation of } \Pi\text{-section in } \text{decibels.}$

The resistors were filed to an accuracy of $\pm 0.05$ ohms or better and then covered with varnish.

Both triggering and time measurements are accomplished by means of crystal-controlled circuits in the A/R Scope. This feature is partly illustrated in Figure 4. Figure 4-a is the trigger which is sent out simultaneously with the start of the sweep (Figure 4-b). The trigger starts the Pulse Former whose output (Figure 4-c) starts the Pulsed Oscillator. The Pulsed
Oscillator generates the signal (Figure 4-d) which is amplified (Figure 4-e) and then produces the sound pulse as described above. Figure 4-f shows the echoes representing reflected sound waves as displayed on the oscilloscope. A delayed sweep circuit in the oscilloscope is used to measure the timing on the leading edge of each echo signal. This feature of the A/R Scope permits an accuracy of 0.02 microseconds. The next section describes in some detail the operations of the electronic circuits which the reader may omit if desired.

**Circuit Details**

In order to determine the time difference between successive pulses with accuracy it is necessary to measure the time of occurrence of the leading edge of each pulse. It is possible to measure the time of occurrence of each pulse with an accuracy of \( \pm 0.02 \) microseconds. The feature of the Du Mont A/R Scope which makes such an accurate measure possible is briefly described. The sinusoidal output of a 100 kc/sec. crystal controlled oscillator is distorted to produce a continuous chain of short one-half microsecond pulses spaced by exactly 10 microseconds. A "Divider" circuit is adjusted to select just one of these pulses once each
1/100 to 1/1000 of a second. This "Divider" circuit allows a choice of a working cycle rate or "pulse recurrence rate" of 100 to 1000 per second, the usual rate being about 300 per second. The selected pulse is then used to synchronize all oscilloscope and external trigger operations for one working cycle. All of the short pulses following the one selected by the divider circuit may be used as markers for time calibration purposes, since they are spaced by exactly 10 microseconds.

The selected pulse, as illustrated in Figure 5-a, is used to trigger a one-shot multivibrator which produces a negative going square wave, (Figure 5-b) of 100 (or 1000) microseconds duration. This square wave form is fed to the input of tube V_2 (Figure 5). Tube V_2 is normally conducting heavily through diode V_1 and R_2 because of the positive grid voltage obtained from positive 250 volts and the grid current IR drop across R_1. The square wave (5-b) voltage cuts off the current through V_2 for 100 microseconds. During this period the condenser C_1 charges linearly through diode V_1 and resistor R_2. Condenser C_1 charges linearly instead of exponentially because V_1 is a constant current diode. The resulting waveform is the linear sawtooth voltage as in Figure 5-c. The cathode of diode V_3, across which the sawtooth voltage is applied, is set at any desired potential by positioning the movable contact of the
potentiometer P. Only that portion of the sawtooth voltage which exceeds the cathode potential can cause diode \( V_3 \) to conduct. This is illustrated by the point \( T \) on the time axis of Figures 5-c, 5-d, 5-e, 5-f, and 5-g. The output of the diode \( V_3 \) is then of the form shown in 5-d. The voltage waveform 5-d is then amplified by later stages, (Figure 5-e) and then differentiated by a short time constant circuit resulting in the peaked waveform Figure 5-f. This voltage is then used to trigger a time base circuit which produces the sawtooth voltage Figure 5-g. This sawtooth voltage may be either 4, 10, or 25 microseconds duration and is applied across the horizontal deflecting plates of the oscilloscope to provide the sweep.

The potentiometer P (Figure 5) has a 10 turn helical winding and the adjustable tap is positioned by a calibrated micrometer "Sweep Delay Dial" having 3600 degrees of rotation. On the 100 microsecond scale the time lapsed between the starting time, say \( t = 0 \), of waveforms (5-b) and (5-c) and until the beginning of the sawtooth voltage (5-g) at time \( = T \), may then be read directly from the Sweep Delay Dial in microseconds to an accuracy of \( \pm 0.02 \) microseconds. (On the 1000 microsecond sweep scale the accuracy is \( \pm 0.2 \) microseconds). The sawtooth sweep voltage (5-g) is of the proper amplitude to cause the time base trace to cover the full four inch
width of the cathode ray tube face on either the 4, 10, or 25 microsecond scales. When the Sweep Delay circuits are not used, the oscilloscope trace may be set for either 100 or 1000 microseconds as shown in Figure 4-f. The above brief explanation shows how it is possible to obtain readings of the time of occurrence of the leading edge of any pulse displayed on the cathode ray tube by positioning the potentiometer with the "Sweep Delay Dial" until the leading edge of the pulse is at the left edge of the oscilloscope trace. High accuracy is obtained because the careful design of the charging circuit, of which Figure 5 is a simplification, makes possible the extremely linear sawtooth voltage of Figure 5-c, and because of the precision design of the helical potentiometer.

Small errors in the readings of the occurrence time of a displayed pulse may occur due to non-linearity of the sawtooth waveform (Figure 5-c) or due to non-linear portions of the helical potentiometer. Errors due to these causes are always very small and adjustment controls for minimizing these errors are incorporated in the A/R Scope. The adjustments are made with the marker pulses, which are generated by the 100 kc/sec. crystal controlled oscillator and associated circuits, displayed on the oscilloscope. Since these pulses are exactly 10 microseconds apart, the error of the "Sweep Delay Dial"
readings may be determined. After proper adjustment the dial readings should not differ from the true time values as indicated by the marker pulses, by more than 0.1 microseconds. One may then make a calibration correction table with the 10, 20, 30, and etc., microsecond intervals, as indicated by the markers, in one column and the "Sweep Delay Dial" readings in a corresponding column. The readings on any displayed sound pulses may then be corrected by reference to this table.

The Du Mont A/R Scope is equipped for synchronizing, displaying and measuring the ultrasonic pulses, as has just been described. The operations of the additional electronic components, as shown in block diagram Figure 3, are now described. The Pulse Former, Radio Frequency Oscillator and Amplifier were built on one chassis from stock radio parts; and the schematic for the combination is shown in Figure 7. This receiver was built around a war surplus radar set intermediate-frequency amplifier which was originally designed for 30 Mc/sec. and had to be redesigned for 10 Mc/sec. Two very stable electronically regulated power supplies were built for these two units.

The operation of the Pulse Former is that of the usual one-shot multivibrator as may be seen by reference to Figure 6. Triode V1 normally conducts
heavily due to the positive bias on its grid obtained from the circuit R₁, R₂, and R₄ (and R₃ for long pulses). Triode V₂ is normally cut off because of the negative grid bias obtained by raising the cathode to a positive potential by the circuit R₇ and R₉. The negative going one microsecond pulse from the A/R Scope is passed through coaxial cable to the Type N coaxial connector N₁. This pulse then cuts off V₁ causing the plate to rise to a higher potential and thus applying a positive pulse through C₂₀ to the grid of V₂. Triode V₂ then conducts through R₉ providing the cathode follower output at N₂ which is a positive going pulse as shown in Figure 4-c. Normally C₂₁ has a negative charge on the V₁ grid side because the plate of V₂ is at a higher positive potential than the tap on the potentiometer R₂. When V₁ is cut off by the one microsecond pulse and V₂ goes conducting the grid of V₁ stays negative until the charge on C₂₁ leaks off through the conducting V₂ and R₂ and R₃. The time required for the charge of C₂₁ to leak off to a value such that the grid potential of V₁ is just above the cut-off value and for V₁ to again conduct, is determined by the setting of R₂. With the switch SW on "pulse width long", an output pulse of 8 to 40 microseconds may be obtained at N₂. On "pulse width short" the output pulse may be varied by R₂ from 1 to 10 microseconds.

The radio frequency circuit operation is as
follows. The positive pulse at $N_2$ is applied to the grid of $V_3$ which is normally cut off. The output of $V_3$ is a negative going pulse which is applied to the grid of $V_4$. Triodes $V_4$ and $V_5$ are normally conducting heavily through the inductance $L_1$. When the negative pulse from $V_3$ cuts off $V_4$, the steady current in $L_1$ is interrupted causing the $L_1C_{10}$ circuit to ring or oscillate. The frequency of this oscillation is variable from 9 to 11 Mc/sec. by adjusting the variable condenser $C_{10}$. The oscillation is sustained for long pulses because the $V_5$ cathode tap on $L_1$ supplies the necessary feedback. As soon as the negative pulse applied to the grid of $V_4$ is over, $V_4$ again conducts and the oscillations of the LC circuit cease. The radio frequency pulse is amplified by the buffer stage $V_6$ and further amplified by $V_7$. The coupling coil $L_4$ provides the low impedance output (about 50 ohms) which is taken off at $N_2$.

The receiver (Figure 7) was originally a 30 Mc/sec. intermediate frequency amplifier having a 2 Mc/sec. band-width. The coils $L_1$ to $L_6$ had to be rewound to tune to 10 Mc/sec. This was accomplished with the aid of a Boonton Q Meter and final alignment accomplished with the aid of a General Radio Company signal generator. Alternate stages were peaked at frequencies on either side of 10 Mc/sec. so that the overall signal voltage output at 9 and 11 Mc/sec. was
about 70% of the output at 10 Mc/sec. This is the usual criteria for a 2 Mc/sec. bandwidth. With a bandwidth of 2 Mc/sec. a one microsecond radio pulse may be passed without distortion of the pulse shape. The ultrasonic signals generated at the quarts were applied at the Type N coaxial connection N₁ and the output from the video amplifier was taken off at N₂. The regulated power supply and the gain control circuits were built on the same chassis. Referring to Figure 8 it is seen that when the potentiometer R₂₄ is varied, from one limit to the other, a variable grid bias ranging from 105 volts negative to 105 volts positive is impressed on V₇. This varies the V₇ conduction current from zero to a value which in turn varies the cathode potential from zero to 100 volts. The plate voltage of stages V₁, V₂ and V₃ is then changed by this circuit and gain control is accomplished.

CONSIDERATIONS IN THE DESIGN OF THE LOW TEMPERATURE APPARATUS

Experimental tests have shown that merely pressing the quartz crystal against the solid specimen does not give an intelligible group of ultrasonic echoes necessary for velocity and attenuation measurements.
The exception to this case is the Hughes method previously mentioned whereby a sound transmitter quartz is pressed against one end of the specimen and the sound receiver quartz against the other. Even with this method attenuation measurements will not be accurate unless the quartz crystals are cemented.

In order to properly convert the energy in the radio frequency pulses into energy in the sound pulse without appreciable pulse distortion, it is necessary that the quartz transducer be cemented to the solid specimen. In the room temperature range the crystal can be attached to the specimen with waxes, salel (phenyl salicylate), stop-cock grease and several other organic agents. To do this the specimen is heated above the melting point of the cement, the cement is applied and the quartz pressed against the specimen until it cools. This results in a very thin film between the crystal and specimen. At the temperature where the film hardens it will have sufficient stiffness to transmit longitudinal and transverse waves into the solid.

If the temperature is lowered too far below the melting point the differential thermal contraction between the film, quartz and specimen may crack the film so that the quartz is no longer attached to the specimen. If the specimen is a fragile solid sufficient stresses may set in to cleave the specimen itself.
The writer, using stop-cock grease as a cement found that X-cut quartz crystals broke away from a beryllium specimen at $136^0\text{K}$ and that Y-cut crystals did this at $112^0\text{K}$. In this case both the metal and the quartz, being stronger than the cement film, were not damaged due to cracking.

J. K. Galt, in measuring longitudinal velocity in a KBr single crystal, used a cement film of pararubber and vaseline. He took the precaution of first cracking the quartz crystal into a mosaic so that each piece could contract individually. Even with this precaution, the stresses set up by the thermal contraction caused the KBr single crystal to crack longitudinally. Fortunately this did not prevent longitudinal velocity measurements which he obtained down to $4.2^0\text{K}$.

Lowering the temperature to about $100^0\text{K}$ causes most materials to go through a large percentage of their overall thermal contraction, since the coefficient of expansion, $\delta$, becomes ever smaller with lowering temperature, and finally $d\delta/dT$ has zero slope at $0^0\text{K}$.
Thus the best solution to the problem seems to be to use a very low melting point cement.

Dr. E. S. Lewis of the Rice Institute Chemistry Department suggested using a mixture of 5 parts ethyl ether, 6 parts isopentane and 2 parts ethyl alcohol as a
cement. This mixture, called EPA, was first used by Dr. Lewis' father, G. N. Lewis and M. Kasha in the study of low temperature chemical reactions. They found that it became a glassy solid at about 100°K.

This EPA mixture was first successfully used by the writer as a cement for ultrasonic measurements in beryllium down to 3°K. In this work procedure was to first cool the specimen to 78°K, remove it from the liquid air, apply a few drops of EPA, wait until the specimen warmed until the solid EPA drops became soft enough and then to press the quartz in position on the drops squeezing the EPA into a thin layer. The electrical contacts were then lowered onto the quartz and the entire assembly was lowered into the liquid air bath. During the process moisture from the air inadvertently got condensed and mixed with the cement. When ultrasonic echoes were obtained the entire apparatus was removed from the liquid air bath and quickly lowered into the experimental chamber of the Rice Institute Collins Helium Cryostat where measurements could be made from 78°K to less than 4.2°K. If the cementing process was not successful, the complicated procedure had to be repeated.

This technique proved successful in longitudinal and transverse velocity measurements in beryllium, in which the transverse velocity was determined down to 3°K and longitudinal velocity to 23°K.
Longitudinal velocity measurements in tin were unsuccessful below 40°K at which temperature the cement layer cracked.

In view of the failure of the other cements even above liquid air temperature and the success of the EPA in the beryllium measurements, it was concluded that the use of EPA offered the best possibilities of success in ultrasonic studies at liquid helium temperatures. Using EPA as a cement, the best results are obtained if it is applied at 120°K. At this temperature it has just the right viscosity so that when the quartz crystal and the solid are pressed together, the viscous cement will be squeezed into a thin layer between the quartz and the solid. Cooling to 100°K solidifies the EPA so that it has sufficient stiffness to transmit both longitudinal and shear waves.

THE LOW TEMPERATURE APPARATUS

Figure 8 shows the construction of the low temperature end of the gear. The dewar flask L surrounds this part and serves as the container for the liquid helium. Dry nitrogen can be let into the dewar vacuum space for precooling.

The vacuum space of dewar L extends 20 inches up from the bottom and the vessel has a single glass wall
the remaining 15 inches to the top of the dewar. The 4 inch I.D. mouth is glass reinforced at the lip and an O-Ring fits into a groove in the glass lip. The dewar is clamped against the bottom of the upper plate H as shown in Figure 3-a. The O-Ring seal makes the system vacuum tight. Two stuffing boxes in the top plate H of Figure 3-a keep the system vacuum tight for up and down motion of the rod G and for rotation of the capillary E.

The EPA cement is applied by letting the liquid flow down the capillary E and out of the nozzle onto the center of the quartz crystal A. The R.F. pulses are fed down the coaxial line made up of stainless steel tube D and the inner copper conductor. The electrical contacts to the quartz are the springloaded plunger C and the stack of steel disk springs B. The framework plates J and K are supported by the thin walled stainless steel tubes D and H. P is sound insulating material holding the specimen in place.

**EXPERIMENTAL PROCEDURE**

The experimental technique of velocity measurement has already been described. The low temperature technique used in velocity measurements in beryllium is described in Appendix II under VELOCITY
MEASUREMENTS.

In preparation for a run at liquid helium temperatures the procedure is as follows: Referring to Figure 8, the quartz crystal A and the specimen are positioned as shown. The interior of the dewar is pumped out and then filled with helium gas to a pressure of one atmosphere. Dry nitrogen gas is then let into the vacuum space of the dewar and the outer dewar I of Figure 3-a is brought up around the inner dewar. The liquid nitrogen in dewar I cools the specimen through the nitrogen exchange gas. When the specimen temperature becomes about $125^\circ K$ the $N_2$ exchange gas is pumped out to allow a slower cooling rate. The EPA cement is then let into the capillary and when the viscous EPA begins to drop out of the nozzle, the capillary E is rotated so that one drop falls in the center of the quartz crystal. The nozzle is then rotated out of the way and the specimen is pushed down onto the quartz crystal. Nitrogen exchange gas is again let into the dewar vacuum space so that the temperature will lower rapidly to about $80^\circ K$.

If the cementing is successful and satisfactory echoes can be obtained, the dewar vacuum space is pumped out to about $2 \times 10^{-5}$ mm of mercury. While this evacuation is taking place the temperature does not rise more than a few degrees above $80^\circ K$. The entire assembly is then removed to the Collins Helium Liquefier and liquid helium
is transferred into the dewar. The temperature of the specimen in the liquid helium range is taken as that indicated by the manometer reading of the vapor pressure of the surrounding helium bath.

EXPERIMENTAL RESULTS

The experimental results are discussed in two groups. The beryllium measurements are discussed in the Appendix. The results of the experiments on tin are discussed in chronological order but separately from those on tantalum.

Run December 15, 1948 On Tin

The tin specimen was made in a vacuum furnace using Bakers C.P. messy tin, having a total impurity content of 0.007 % of As, Pb, Fe, Zn and Cu. The tin was then made into a cylinder 2.5 cm in diameter and 5.047 cm long. An X-cut quartz crystal was cemented to the tin in the same manner previously described for the beryllium. The longitudinal velocity measurements are given in the following table.
<table>
<thead>
<tr>
<th>Temperature K</th>
<th>Longitudinal Velocity cm/sec $\times 10^{-5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>$3.35 \pm 0.04$</td>
</tr>
<tr>
<td>118</td>
<td>$3.48 \pm 0.04$</td>
</tr>
<tr>
<td>114</td>
<td>$3.48 \pm 0.04$</td>
</tr>
<tr>
<td>113</td>
<td>$3.51 \pm 0.04$</td>
</tr>
<tr>
<td>108</td>
<td>$3.51 \pm 0.04$</td>
</tr>
<tr>
<td>105</td>
<td>$3.50 \pm 0.04$</td>
</tr>
<tr>
<td>92</td>
<td>$3.48 \pm 0.04$</td>
</tr>
<tr>
<td>90</td>
<td>$3.47 \pm 0.04$</td>
</tr>
<tr>
<td>84</td>
<td>$3.45 \pm 0.04$</td>
</tr>
<tr>
<td>71</td>
<td>$3.51 \pm 0.04$</td>
</tr>
</tbody>
</table>

These velocities were measured by timing the leading edge of three echoes and averaging the time interval between them. The third echo signal to noise ratio was only about three so it was difficult to measure its leading edge accurately. The error in these velocity measurements is a little worse than 1%. In comparison, the signal to noise ratio of the eighth echo in the beryllium measurements was about 15 and its leading edge could be very accurately timed. The beryllium measurements yielded longitudinal velocities accurate to about 0.05%. The measurements were not carried below $70^\circ$K because of liquefier trouble.

Run February 15, 1949 On Tin

The experimental apparatus was made ready and echos were obtained at liquid air temperatures. A broken clamp on the little engine of the liquefier prevented low temperature measurements. The tin specimen was used for another experiment.
Run August 25, 1949 On Tin

On this run, the time required for the sound pulses produced at one end of the tin specimen to travel through it to the other end was measured. This is the Hughes technique, already described, wherein X-cut quartz crystals are held against the flat ends of the specimen by a small pressure. The specimen was a tin rod 7.719 cm. long and 1.4288 cm. diameter, made from Bakers C.P. mossy tin which had been melted in vacuo. One half inch X-cut quartz crystals were pressed against the ends of the rod (without cement), with phosphor bronze spring contacts. A 300 turn coil of Number 30 copper wire was wound on a 3/4 inch form around the specimen. This winding was connected to a ballistic galvanometer in order to indicate the superconducting transition. A second winding of 600 turns Number 30 copper wire wound on a 1-1/4 inch form around the first. This coil produced a field of 100 gauss per ampere at the center of the specimen and could be used to destroy superconductivity.

This assembly was mounted on a support and placed in the experimental chamber of the Helium cryostat. Temperatures in the liquid helium range were measured with a vapor pressure thermometer. The bulb of this thermometer was located next to the assembly.

A longitudinal pulse travels straight through
the rod in time $t_1 = \frac{1}{\frac{1}{v_1}} + t_o$. Where $l$ is the length of
the rod, $v_1$ the longitudinal velocity and $t_o$ is the
conversion time for the sound pulse to be generated at
one end and detected at the other. The part of the
longitudinal wave grazing the surface is converted into
a transverse wave which travels across the rod at an
angle $\theta$ with respect to the normal to the cylindrical
surface with $\sin \theta = v_t / v_1$. The delayed pulse due to
this phenomena arrives at a time $\Delta t_{21}$ later than $t_1$ where

$$\Delta t_{21} = \frac{D \left[ \frac{v_2^2 - v_t^2}{v_2} \right]^{1/2}}{v_2 v_t}$$

A pulse twice reflected across the rod arrives still later

$$\Delta t_{31} = \frac{2D \left[ \frac{v_2^2 - v_t^2}{v_2} \right]^{1/2}}{v_2 v_t}$$

These pulses could be timed on the oscilloscope
to $\pm 0.02$ microseconds, if the pulse to noise ratio was
good enough, and if the leading edge of the pulse was
steep enough to be distinguished from noise fluctuations.
Measurements were taken in the liquid air and liquid
helium ranges. Some of the data are tabulated below.

<table>
<thead>
<tr>
<th>Temperature °K</th>
<th>First Arrival</th>
<th>Second Arrival</th>
<th>$\Delta t_{21}$</th>
<th>Third Arrival</th>
<th>$\Delta t_{31}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>79</td>
<td>23.90</td>
<td>31.87</td>
<td>7.97</td>
<td>34.80</td>
<td>10.90</td>
</tr>
<tr>
<td>4.2</td>
<td>24.20</td>
<td></td>
<td></td>
<td>36.49</td>
<td>12.49</td>
</tr>
<tr>
<td>3.77</td>
<td>24.00</td>
<td></td>
<td></td>
<td>36.75</td>
<td>12.75</td>
</tr>
<tr>
<td>3.31</td>
<td>23.90</td>
<td></td>
<td></td>
<td>36.50</td>
<td>12.60</td>
</tr>
</tbody>
</table>

In both the liquid air and liquid helium ranges
it was observed that when current was sent through the
field coil, the position of the pulses on the oscilloscope would shift slightly, and change in amplitude. The shift was a decrease in time of arrival of 0.02 to 0.03 microseconds. At the same time the signal increased very slightly in amplitude. With zero field current, disconnecting the ballistic galvanometer gave the same effect. Again the same effect was observed when the specimen became superconducting. So the jump in amplitude and shift in leading edge position could not be definitely tracked down. Pumping also caused erratic shifting of the echoes over a few hundredths of a microsecond. The cause of the effect was assumed due to a change in the impedance of the circuit through which the R.F. currents flowed to the quartz crystals. Such a change accounts for a shift in signal amplitude. The shift in time is due to the shift in amplitude because the leading edge of the pulses is not vertical but sloping.

Due to the uncertainties in this experiment it cannot be said that there is a shift in the velocity or attenuation of the 10 Mc/sec sound when the tin goes superconducting. However the transverse velocity can be calculated and the elastic moduli obtained from these results. For example at 79°K

\[
\frac{\Delta t_{31}}{2D} = \frac{10.90 \times 10^{-6} \times 3.50 \times 10^5 \times v_t}{2 \times 1.4288} = \left[\frac{(3.50 \times 10^5)^2 - v_t^2}{2}ight]
\]

\[
v_t = 2.10 \times 10^5 \text{ cm/sec}
\]
The shear modulus is then, with density taken as 7.3

\[
\mu = \rho v_t^2 \\
= 3.21 \times 10^{11} \text{ dynes/cm}^2
\]

and the bulk modulus

\[
k_s = \rho \left( v_1^2 - \frac{4}{3} v_t^2 \right) \\
= 4.65 \times 10^{10} \text{ dynes/cm}^2
\]

This is about the same as Grüneisen's isothermal value of \(4.67 \times 10^{11}\) dynes/cm.² at \(-190^\circ C\). (His measurement of isothermal compressibility was \(210 \times 10^{-6} \text{ mm}^2/\text{kg}\)).

Run October 5, 1949 On Tin

The Hughes technique was also used on this Run.

The R.F. connections to the crystals were arranged so that R.F. currents could not flow through the specimen and the ballistic galvanometer and field coils were not used. No change was observed on passing through the superconducting transition although electronic troubles prevented a thorough investigation.

Run January 6, 1950 On Tin

This was the first Run, done outside of the helium cryostat using the apparatus described under LOW TEMPERATURE APPARATUS. The tin specimen was one inch in diameter and 16.198 mm. long.

The room temperature longitudinal velocity,
averaged over 8 echos was $3.340 \pm 0.003 \times 10^5 \text{cm/sec.}$
At $100^\circ\text{K}$, an average over 3 echos gave a value of
$3.48 \pm 0.04 \times 10^5 \text{cm/sec.}$ Measurements in the liquid
helium range were unsuccessful.

**Run January 20, 1950 On Tin**

The one inch diameter tin cylinder used in the
December 15, 1948 run was cut down to 3.829 cm. The
procedure discussed above, for low temperature runs in
the external dewar flask, was followed. The quartz was
cemented to the tin specimen at $120^\circ\text{K.}$ Figure 9-a and
Figure 9-b (with reduced amplifier gain) are
reproductions of the oscilloscope trace observed at $85^\circ\text{K.}$
The longitudinal velocity, averaged over the first 3
echos was $3.48 \pm 0.05 \times 10^5 \text{cm/sec.}$ The large error is
due to error in locating the leading edges of the second
and third echos.

The attenuation was measured by switching in
sections of the $\Pi$-network attenuator. The first echo
required 28 decibels to reduce it to noise while the
second echo was reduced to noise level with 14 decibels.
The uncertainty in the 14 decibel difference is $\pm 2\text{db.}$
The attenuation for the round trip through the tin
specimen is $\frac{14 \pm 2}{2 \times 3.829}$ or $1.8 \pm 0.3 \text{ decibels per}
\text{centimeter.}$ The pressure attenuation of the sound wave
is described by $p = p_0 e^{-\alpha x}$
where, at \(85^\circ K\) in the tin specimen used,

\[
\alpha = \frac{1.8 + 0.3}{20 \times 0.4343} = 0.21 + 0.03 \text{ per cm.}
\]

Figure 9-c shows the oscilloscope trace after the temperature had been lowered to the liquid helium range and later permitted to warm back to \(85^\circ K\).

On transferring liquid helium into the dewar the different thermal contractions of the quartz, EPA cement film, and the tin specimen set up sufficient stresses to break the quartz-specimen bond. This breaking had occurred on previous runs in the helium cryostat at about \(40^\circ K\). As a result, the echoes of Figure 9-a could not be obtained in the liquid helium range. The resulting narrowed acoustic bandwidth required a 12 microsecond pulse at \(4.2^\circ K\) to obtain the oscilloscope trace shown in Figure 9-d. Inserting 10db at \(4.2^\circ K\) gave the oscilloscope trace in Figure 9-e.

The amplitude of the signal in Figure 9-d was caused to fluctuate by sudden pressure changes or rapid pumping. If the temperature was lowered slowly, the echo remained unchanged until the superconducting transition was reached. When the temperature was lowered to \(3.7^\circ K\) the echo suddenly increased in amplitude to that shown in Figure 9-f. Figure 9-g shows the signal at \(3.7^\circ K\) reduced by 10 db.

The amplitude of the signal increased further
when the temperature was lowered to 3.48°K, Figure 9-h and still further at 3.38°K. The second echo was concealed by the receiver swamping. The third echo began to appear at 3.48°K as shown in Figure 9-h and became stronger as shown in Figure 9-i. Figure 9-k shows the signals at 3.38°K attenuated by 10 db. When the pumping was stopped the echoes reverted to that of Figure 9-d even though the temperature, indicated by the vapor pressure, was below 3.7.

At first it was thought that these data indicated a shift in the ultrasonic attenuation at the transition temperature, and 36 photographs of the oscilloscope trace were taken between 4.2°K and 3°K in order to determine the magnitude of the attenuation change. The possibility of amplitude changes with pressure fluctuations had not been taken into account. Also the possibility of changes in the driving impedance due to changes in circuit conditions when the specimen goes superconducting had not been weighted. Later tests at room temperature showed a similar variation of signal amplitude with pressure changes for a transducer improperly bonded to the specimen. These considerations indicate that the amplitude changes observed in this experiment cannot be attributed to a change in the ultrasonic attenuation when the specimen goes superconducting.
Run April 13, 1950 On Tin

Using the same tin specimen (3.829 cm. long, one inch in diameter) as on the January 20, 1950 run. The longitudinal velocity measured at 80°K was 3.50 ± 0.03 x 10^5 cm/sec. The attenuation averaged over 3 strong echoes was 15 ± 1 decibels per 7.658 cm. This is in agreement with previous measurements.

When liquid helium was transferred the first echo could not be seen for it was obscured in the receiver recovery.

The second and third echoes were observed at about 46 microseconds and 67 microseconds respectively and the signal to noise ratio was respectively about 5 to 1 and 2 to 1. The pulse width was set at 6 microseconds. Although the pulse shape was fairly good, the leading edge of the second echo could not be located more accurately than about 0.4 microseconds. The time location, time interval between the two pulses and the accuracy with which the third echo could be located (0.8 to 1 microsecond) prove that the 10Mc/sec. sound velocity at 4.2°K does not differ by more than about 1.6% from the sound velocity at 80°K.

The amplitudes of these echoes did not fluctuate with pumping. Attenuation measurements were made by switching in sections of the $\mathcal{N}$-network attenuator until
the echo amplitudes were just distinguishable in the noise. The measurements are given in the following table.

<table>
<thead>
<tr>
<th>$T^\circ K$</th>
<th>Second Echo db above noise</th>
<th>Third Echo db above noise</th>
<th>Attenuation db per 7.658 cm.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.11</td>
<td>$18 \pm 1$</td>
<td>$12 \pm 2$</td>
<td>6</td>
</tr>
<tr>
<td>3.36</td>
<td>$17 \pm 1$</td>
<td>$8 \pm 1$</td>
<td>9</td>
</tr>
<tr>
<td>3.39</td>
<td>$18 \pm 1$</td>
<td>$11 \pm 2$</td>
<td>7</td>
</tr>
<tr>
<td>3.67</td>
<td>$20 \pm 1$</td>
<td>$10 \pm 1$</td>
<td>10</td>
</tr>
<tr>
<td>3.84</td>
<td>$20 \pm 1$</td>
<td>$8 \pm 1$</td>
<td>12</td>
</tr>
<tr>
<td>4.05</td>
<td>$18 \pm 1$</td>
<td>$8 \pm 1$</td>
<td>10</td>
</tr>
<tr>
<td>4.20</td>
<td>$18 \pm 1$</td>
<td>$6 \pm 1$</td>
<td>12</td>
</tr>
<tr>
<td>10</td>
<td>$20 \pm 1$</td>
<td>$10 \pm 1$</td>
<td>10</td>
</tr>
<tr>
<td>14</td>
<td>$20 \pm 1$</td>
<td>$10 \pm 2$</td>
<td>10</td>
</tr>
<tr>
<td>20</td>
<td>$20 \pm 1$</td>
<td>$5 \pm 1$</td>
<td>15</td>
</tr>
<tr>
<td>31</td>
<td>$20 \pm 1$</td>
<td>$5 \pm 1$</td>
<td>15</td>
</tr>
<tr>
<td>64</td>
<td>$20 \pm 1$</td>
<td>$5 \pm 1$</td>
<td>15</td>
</tr>
<tr>
<td>82</td>
<td>$20 \pm 1$</td>
<td>$5 \pm 1$</td>
<td>15</td>
</tr>
</tbody>
</table>

Below 3.67$^\circ$K the number four echo was just distinguishable in the noise. This indicates that the attenuation is decreasing with temperature.

**Run April 25, 1950 On Tin**

The tin specimen 3.829 cm was again used in this run. After preparation and obtaining signals at 80$^\circ$K, measurements were made of the driving point impedance of the transducer. The Boonton Q-Meter was
used for this purpose.

Actually the Q-Meter is not very satisfactory for measurements of the effective load resistance of the transducer for pulses since the C-W output of the Q-Meter causes the tin specimen to act as an acoustic resonator. The Q-Meter connections were made at the coaxial terminal at the top of the apparatus. Normally the output of the impedance matching network is connected to this terminal. The coaxial line to the transducer is the 35 inch stainless steel tube that supports the low temperature end of the gear with a copper lead inside. \((Z_0 = 138\ \text{ohms})\). The admittance at the point of connection is \(Y = G - jB\). The value \(R = \frac{1}{G} \) is calculated for this is the effective parallel load resistance at the driving point. The coaxial line is essentially a parallel capacitance that can be balanced out by the inductance of the matching network when pulses are used. The 35 inch length of the line is less than \(\frac{1}{30}\) of the wavelength at 10 Mc/sec, and so no important corrections are necessary for these measurements. The off-resonant reactance will be somewhat affected by the line but the resistive part is essentially the load resistance of the quartz transducer. Figure 10-a shows the effective load resistance vs frequency for the tin specimen at 80°C.

After the liquid helium was transferred signals could not be obtained unless the specimen was pressed
against the quartz crystal. This can be done from the outside by a downward force on the rod G of Figure 8.

The first three echoes could be observed but pumping caused erratic amplitude fluctuations so that attenuation measurements could not be made. Q-Meter measurements of the load resistance were made at 4.2 and at 2.5°K. These are shown in Figure 10-b and 10-c respectively. The best echoes were obtained at 80°K with the frequency set at 10.4 Mc/sec. At 4.2°K, however, the strains set in by thermal contraction on cooling causes the EPA cement film to crack. This poor bonding of the quartz to the specimen results in a reduction in acoustic loading, (Conclusión), a decrease to 9.9 Mc/sec. in the resonant frequency of the transducer, and a reduction in the band pass of the transducer. As in several previous experiments it then becomes necessary to broaden the output pulse of the transmitter in order to obtain echoes.

Run August 10, 1949 On Tantalum

This experiment was done in the experimental chamber of the helium cryostat, using the same procedure for cementing the quartz as described for the December 15, 1948 Run on tin. The specimen was a one inch diameter rolled tantalum rod 2.52 cm. long. No echoes could be obtained in the specimen until it had been heat
treated by heating in vacuo to about 1100°F for an hour. After this treatment some echoes could be obtained although they were badly distorted by the inhomogeneity of the specimen.

At 4.2°K it was not possible to obtain the usual echo pattern as in Figure 1-c. The pulse width was increased to about 10 microseconds in order to obtain echoes. The first echo at about 12 microseconds had a poorly defined leading edge and could not be measured accurately. No other echoes could be definitely identified as being repetitions of this first one at 12 microseconds, except possibly the Number 10 echo at 102 microseconds. This signal was watched as the temperature was raised from 4.2°K to the tantalum transition at 4.38°K. The slight decrease in amplitude of the echo at 102 microseconds at the transition was not consistent with each increase or decrease of the temperature through the transition. Multiply reflected and scattered signals could be observed out to 900 microseconds showing that the absorption of the sound energy was very small. The effect of shifts in the transducer output with pressure changes was observed.

**Run May 7, 1950 On Tantalum**

Velocity measurements were made in a tantalum specimen 3.849 cm. long and one inch in diameter. This
specimen had not been heat treated. Readable signals could not be observed at room temperature. Readable signals were observed at 80°K using EPA to cement the quartz crystal. The leading edges of the echos at 80°K were badly distorted and echos from inhomogeneities were observed. The round trip travel time averaged over four echos was 17.1 microseconds. This corresponds to a longitudinal velocity of \( 4.5 \times 10^5 \) cm/sec.

The heat treated tantalum specimen length 4.201 cm. was used on the low temperature run. Figure 10-a shows the oscilloscope trace at 4.2°K, below the superconducting transition (4.38°K). Figure 10-b shows the trace at 4.45°K, above the transition. While the temperature was lowered through the transition no change in position or height of the echos could be observed on the 100 microsecond sweep.

Figure 10-c shows the scope trace for tantalum at 4.2°K on the 1000 microsecond sweep. The multiply reflected and scattered pulses can be seen out to 1000 microseconds. No change was observed in the amplitude of the echos on cooling or warming through the transition. Figure 10-d shows the oscilloscope trace at 4.2°K on the 1000 microsecond sweep in which the pulse width was increased to 10 microseconds. Again there was no noticeable change in the amplitude of the echos over this range when the temperature was raised or lowered through
the transition. It is seen that tantalum is an excellent material for propagating sound pulses and there is very little attenuation at low temperatures. Very accurate measurements of the moduli and velocities could be obtained in a sufficiently homogeneous specimen.

CONCLUSIONS

Figure 2-a indicates that, for tin, there would be a 31% decrease in the sound attenuation on lowering the temperature through the superconducting transition if all the attenuation is due to thermal conduction. Reference to Table III (page 41) shows that this is just about the magnitude of the experimental error of the attenuation measurements on the April 13, 1950 Run. Thus a change in the sound absorption could occur without making itself apparent. Assuming that the 10 db attenuation at 4.2°K is due only to the mechanism of thermal conduction we have

\[ \alpha = \frac{10 \text{db}}{7.658 \text{ cm. x 8.68}} = 0.015 \text{ per cm.} \]

which places an upper limit on the value of \( \frac{C_p}{C_v} - 1 \);

\[ \frac{\alpha_{\text{th}}}{\gamma - 1} = 3200 \text{ cm.}^{-1} = \frac{1.5 \times 10^{-2}}{\gamma - 1} \text{ cm.}^{-1} \]
\[
\frac{C_p}{C_v} - 1 \lesssim 4.5 \times 10^{-6} \text{ at } 4.2^\circ K
\]

provided that the classical theory expression for \( \chi \) thermal which holds for gases can be proved to hold also for metals.

An interesting result of the April 13, 1950 Run is the decrease in the attenuation of 10Mc/sec. sound in tin with decreasing temperature below 80^\circ K. (Table III, page 41). These data have not yet been correlated with one of the contributing factors, i.e., attenuation due to Raleigh scattering from grains since the grain structure has not been determined. Experimental measurements of attenuation at different frequencies should be made so that the \( f^2 \) dependence of \( \chi \) thermal and \( \lambda^{-4} \) dependence of attenuation due to scattering can be separated.

Adequate experimental methods for precision measurements of high frequency sound attenuation in metals at low temperatures have yet to be developed. Low frequency techniques (up to 50 kc/sec.) are very accurate, but the ultrasonic pulse method used in the present experiments is unsatisfactory for precision attenuation studies in metals although it is the most precise tool known for studying sound velocities. In liquids and gases, however, very accurate measurements of the attenuation can be made since other factors aiding this
accuracy are controllable.

The discontinuous change, previously reported by the writer, of the attenuation coefficient for tin, is now known to have been a false conclusion. This was based on the effects illustrated by photographs 9-d to 9-k, taken during the January 20, 1950 Run. The extraneous effects had not been properly considered. For this mistake the author humbly apologizes.

The results obtained in tantalum also do not show a discontinuous change in the sound attenuation at the transition. However they do show that the overall attenuation is very small. The EPA cementing technique works as well for tantalum as for beryllium and if a single crystal or a homogeneous specimen of tantalum could be obtained, very accurate measurements of the velocity could be made at low temperatures.

Using the Hughes technique, (page 6 and page 33), the elastic properties of solids can be readily studied, over the temperature range of the Helium Cryostat (300°K to 4.2°K).
The curves in Figure 10 show the effective parallel resistance, as a function of frequency, of the quartz transducer radiating sound into the tin specimen. The measurements of this resistance were made with a Q-meter using its continuous wave output.

This resulted in the specimen behaving as an acoustic resonator. Had the sound attenuation in the tin specimen been large enough so that an inappreciable amount of energy reflected and returned to the quartz, then the curves of Figure 10 would represent the transducer characteristics for pulse or one-way continuous wave transmission. However, since the attenuation in tin is fairly large, the curves in Figure 10 are sufficiently representative to permit conclusions as to the changing behavior, with the temperature of the transducer for one-way transmission and reception. The conclusions that may be drawn are enhanced by correlation with the photographs of the pulses on the oscilloscope trace, since it is well known that the bandwidth of the transducer is inversely proportional to the width of the pulse that can be transmitted without distortion.

In Figure 10-a the bandwidth at 80°C is seen to be about 0.8 Mc/sec. A pulse width, \( \delta = \frac{2}{\Delta} = 2.5 \) microseconds, could be transmitted without distortion. In Figure 9-a, the pulse width was 5 microseconds and the
shapes of the echos are essentially that of the transmitted pulse which was observed by displaying it alone on the oscilloscope. It will be noted, however, that the center frequency of the transducer is 10.4 Mc/sec. The acoustic loading, at 800K, was quite satisfactory for transmission of longitudinal pulses and for longitudinal velocity measurements, accurate to about one percent and attenuation measurements accurate to about 20% by means of the attenuator "C" shown in Figure 3-a.

The curves of Figure 10 represent a variety of experimental conditions. The characteristics of the quartz-to-solid cement bond is different in each case. For the case of Figure 10-b at 4.20K, echos could not be observed. The application of a downward external pressure on shaft "G" of Figure 8, caused the transducer and the solid to be pressed hard together and the transducer to be pressed hard against the concentric electrical contacts. However, the curve of effective parallel resistance versus frequency of Figure 10-b at 4.20K is only slightly different from that of Figure 10-c (2.50K with pressure). They have the same resonance frequency, 9.9 Mc/sec. although Figure 10-c is slightly wider. Yet pulses could be received and attenuations measured (to about 30%) in the case of Figure 10-c. Thus the application of external
pressure aided in the pulse reception. The echoes obtained were much weaker than those shown in Figure 9-a. This indicates either that the EPA cement film was cracked so badly that pulses could not be efficiently transmitted or that lateral stresses across the face of the quartz crystal greatly reduced its efficiency. One would be inclined to think that the efficiency of the quartz is effected by both factors. Another alternative is that the EPA cement film contraction causes it to leave gaps in the cemented interface so that the quartz is not in contact with the tin specimen over the whole area of the quartz. This would reduce the radiated sound power and the overall efficiency of the transducer. Yet the application of pressure changed the shape of the curve slightly and permitted the reception of echos.

The resonant frequency of the transducer was lowered from 10.4 Mc/sec. at 80°K to 9.9 Mc/sec. at 4.2°K and 2.5°K. This cannot be explained on the basis of a reduced acoustic loading, from the viewpoint of radiating into a media of lowered acoustic resistance. A mathematical analysis by Roth\(^{18}\), of an infinite quartz slab coated on each face with thin metal films, with each film being at the same R.F. potential over the film surface, leads to the following conclusions. Note first that a slab of quartz some 50 wavelengths across is sufficiently
described by his analysis and that the cement film holding the quartz to the solid specimen should have the same acoustic resistance, \( \rho v \), as the solid to which the quartz is cemented. Roth's conclusions for this ideal case are:

1) The bandwidth increases as the load impedance increases. The bandwidth here also means the electrical bandwidth or the half-width of the resistance versus frequency curve where the resistance is either the parallel or series part of the electrical impedance at the driving point.

2) The maximum acoustic power output at resonance, for a given R.F. driving source varies inversely to the bandwidth of the transducer.

3) With a high internal impedance source, the resonance frequencies decrease with an increase in loading; but with a low internal impedance driving source the resonance frequencies remain very nearly constant.

4) Loading by materials having impedances greater than that of the transducer leads to doubly peaked resonance curves.

The behavior of the transducer at low temperatures due to contraction of the EPA cement film, the quartz, and the solid, together with the possibility of lateral stresses across the quartz face due to contraction of the cement, would be expected to deviate somewhat from the "ideal" case.
above. However, a comparison against the "ideal" conclusions is indicated.

The half-width of the R.F. resistance versus frequency would be expected to increase if, for example, the transducer, radiating sound into liquid helium was then caused to transmit sound into carbon tetrachloride where the acoustic resistance $\rho v$ is much greater than that of liquid helium. However, in the present comparison the solid media was always tin. The reduced bandwidth in the case of Figures 10-b and 10-c from that of Figure 10-a suggest that the effective acoustic impedance of the EPA film-tin solid media was considerably reduced at 4.2°K from that at 80°K. Certainly the efficiency of the transducer was reduced. If the mechanical contact between the quartz and the tin specimen is not continuous at the interface then the displacement of the tin surface at the interface would not be expected to follow very well the mechanical vibration of the transducer. During the rarefactions, at the interface, of the vibrations, the quartz would be essentially unloaded whereas during the compressions of the sound, the quartz would be loaded. The average acoustic loading would be expected to be lowered. This would also explain why pressing the quartz against the tin specimen, Figure 10-c, gave stronger echos and a change in the shape of the bandwidth curve. Then, contrary to
conclusion (3), the reduced acoustic loading at 4.2°K and 2.5°K, from that at 80°K, resulted in a decrease in the resonant frequency. This is also contrary to the results of Pellam and Squire⁵. Their Q-meter measurements of the effective parallel radiation resistance gave a bandwidth of 1.7 Mc/sec. and a center frequency of 15 Mc/sec. for liquid CCl₄ and a bandwidth of 0.2 Mc/sec. at a center of frequency of 15.3 Mc/sec. for liquid helium. Their results of course agree with conclusion (3) since the acoustic loading for liquid helium was less than that of the CCl₄. However, in this case, the reduction in acoustic loading was not due to radiation into a different solid but that of an unloaded crystal. Cracking of the EPA cement layer caused the crystal to become essentially unloaded at 4.2°K.

When the tin specimen was allowed to warm back up to 80°K, Q-meter measurements gave the same bandwidth and center frequency (9.9 Mc/sec.) as at 4.2°K. However, the efficiency was in general much better, and much stronger echos were observed than at 4.2°K. Of course the echos were not those of Figure 9-a, but were as shown in Figure 9-c. The stronger echos began to appear, during the warm-up, at about 60°K. This is strongly indicative of a reduction in lateral stresses across the quartz due to differential thermal contraction.

The accumulated experimental evidence obtained
during the course of this low temperature research on ultrasonic propagation in solids leads to these conclusions:

a) The use of EPA cement as a quartz-to-solid bond is, at this time, the only known solution to the problem of echo ranging in metallic solids at temperatures considerably less than 800K. In beryllium the use of EPA cement was successful at 30K, while in tin it was successfully used to about 300K, at which temperature the differential thermal contraction became excessive.

b) After excessive thermal contraction causes cracking of the EPA cement film, there is a reduction in acoustic loading and a decrease in the resonance frequency.

c) If the cracking of the cement film occurs at the interface between the film and the solid, but the film remains cemented to the quartz, then the differential contraction between the film and the quartz sets up lateral stresses across the quartz at the interface, sufficient at low temperatures to lower considerably, the efficiency of the transducer.

d) For a given solid the bandwidth increases with the viscosity of the cement, becoming very broad (1 to 2 Mc/sec.) as the cement becomes solid. The resonance frequency of the bandwidth curve increases with
the viscosity of the cement.
ACKNOWLEDGEMENT

The author wishes to acknowledge his indebtedness to Dr. C. F. Squire for his encouragement and assistance during this investigation. The author also wishes to thank Mr. R. W. Schmitt, Mr. A. Lathrop, Mr. P. B. Alers, Mr. L. Morton, Mr. Claude Grenier and Mr. N. Meunch who were very helpful, from time to time, in the production of the liquid helium for these experiments.
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10) A. Radmakers, Physica, 15, 849 (Oct. 1949)
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APPENDIX I

The composite Oscillator


The electrical reactance of the oscillator varies critically in the neighborhood of resonance at which radio frequency, the amplitude of the mechanical vibrations will be a maximum. This resonance is indicated by a minimum in the R.F. driving current of the power oscillator. The resonance frequency \( f = f_2 \) of the quartz is first obtained. The specimen is then cemented to the quartz and the resonance frequency \( f \) of the composite oscillator is measured. The equation

\[
\frac{M_2}{M_1 y_2} \tan y_2 = \frac{1}{y_1} \tan y_1 \quad (1)
\]

is then solved for one of the resonance modes of longitudinal vibration of the specimen.

For the quartz

\[
y_2 = \pi \frac{f}{f_2} \left[ 1 - \frac{\pi^2 \rho_z^2 \Theta_z^2}{2A L_z^2} \left( 1 - \frac{f_2}{f_1} \right) \right]
\]
and for the specimen

\[
y_1 = \frac{\pi f}{f_1} \left[ 1 - \frac{\pi^2 \sigma_i^2 \Theta}{2A \ell_i^2} \right] \left( 1 - \frac{f}{f_i^2} \right)^2
\]  

(3)

where

\( \sigma_i^2 \) = Poisson's Ratio
\( \Theta_i^2 \) = Moment of inertia about the Z-axis
\( A \) = cross section of the rods
\( \ell \) = length.

In equation (2) all the quantities in the right hand side are known so that \( \frac{M_2 \tan y_2}{M_1 y_2} = W \) is determined.

Then

\[
W y_1 = W \frac{\pi f}{f_1} \left[ 1 - C \left( 1 - \frac{f}{f_i^2} \right) \right] = -\tan \left\{ \frac{\pi f}{f_1} \left[ 1 - C \left( 1 - \frac{f}{f_i^2} \right) \right] \right\}
\]

is solved for \( f_1 \).

The Youngs modulus \( G_1 \) is calculated from

\[
f_1 = \frac{m}{2 \ell_i} \sqrt{\frac{G_i}{\rho_i}} \left[ 1 - \frac{\pi^2 n^2 \sigma_i^2 \Theta_i}{4A \ell_i^2} \right]
\]  

(4)

The approximate value of \( \sigma_i \) is needed in order to obtain the Youngs modulus. This can be obtained from independent sources.


As discussed on page 6, the time delay between the longitudinal pulse that travels through the specimen
and the pulses reflected from the surface, is measured on the oscilloscope.

The time of arrival \( t_1 \) of the direct pulse will be

\[
t_1 = \frac{d_1 + d_2 + d_3 + d_4 + d_5}{V_1} = \frac{1}{V_1}
\]

Where \( V_1 \) is the longitudinal velocity. At the surface the longitudinal wave is converted into a transverse wave which travels with velocity \( V_t \) across the rod at an angle \( \theta \) with respect to the normal

\[
\sin \theta = \frac{V_t}{V_1}
\]

This pulse arrives at a later time \( t_2 \).

\[
t_2 = \frac{d_1 + d_3 + d_4 + d_5}{V_1} + \frac{D}{V_t \cos \theta}
\]

while \( t_1 \) can be written

\[
t_1 = \frac{d_1 + d_3 + d_4 + d_5}{V_1} + \frac{D \tan \theta}{V_1}
\]

The time delay measured on the oscilloscope will then be
\[ \Delta t_{21} = \frac{D}{v_t \cos \theta} - \frac{D \tan \theta}{v_e} \]

Substituting \( \sin \theta = \frac{v_t}{v_1} \) gives

\[ \Delta t_{21} = D \left[ \frac{v_e^2 - v_t^2}{v_e v_t} \right]^{1/2} \]

A twice reflected pulse will arrive at a still later time \( t_3 \).

\[ \Delta t_{31} = \frac{2D}{v_t \cos \theta} - \frac{2D \tan \theta}{v_e} \]

The delay time will be

\[ \Delta t_{31} = 2D \left[ \frac{v_e^2 - v_t^2}{v_e v_t} \right]^{1/2} \]

Measurement of the delay times for rods of several lengths and thicknesses permits an accurate determination of \( v_1 \), \( v_t \) from which the elastic moduli can be calculated.
Ultrasonic Measurements in Metallic Beryllium at Low Temperatures*

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The Rice Institute, Houston, Texas
(Received June 9, 1949)

Using the ultrasonic pulse method, the velocity of longitudinal waves in metallic beryllium has been measured from 300°K to 23°K, and transverse waves from 300°K to 3°K. The adiabatic moduli and compressibility have been computed from the velocities. A new technique for affixing the quartz transducer to the solid for ultrasonic measurements, in suitable solids, below 100°K, has been successfully employed for the measurements in beryllium.

INTRODUCTION

The ultrasonic pulse method described by Pellam and Squire for sound ranging in liquid helium and by Huntington for velocity measurements in solids has been applied to measure the longitudinal and transverse velocities of 10 Mc/sec. sound pulses in metallic beryllium. The usual technique is to apply the electrical pulse from a transmitter to a quartz crystal which is rigidly affixed to the solid sample. The quartz transducer then generates a sound pulse which is propagated through the material, reflects from the surface opposite the quartz and then returns to produce an echo. Direct measurements are made on the echoes of the pulses displayed on an oscilloscope such as the DuMont A/R 256-D. The velocities are determined from the round trip distance traveled by the sound pulse and the timing of the corresponding echoes.

The selection of beryllium as the initial material of a series of proposed echo ranging experiments was guided by several factors, one of which was to develop a material for affixing the quartz crystal to the solid that would hold at low temperatures. These compounds and techniques of application could then be used in the measurements on the more fragile materials which might be damaged by thermal contraction at the low temperatures. In addition beryllium has certain interesting properties. Simon and Critescu have observed a hump in the specific heat between 10°K and 14°K. Although the specific heat is small in that range, the hump is about twice the value one would obtain if an extrapolation, according to the Debye T^3 law, were made from about 18°K to 0°K. Simon and Critescu attribute the hump to electronic causes; however Squire has measured the resistivity of beryllium and observed no anomaly in the 10 to 14°K region. The resistivity curve in itself is peculiar in that it does not approach zero near the absolute zero, as do most ordinary metals, but levels off to about three-tenths of the room temperature value. Because of the specific heat hump one might expect a slight change in the ultrasonic attenuation and possibly a measurable change in the velocity.

The difficulties in making ultrasonic measurements in solids at low temperatures are mainly due to the different thermal contraction rates of the quartz crystal and the solid. The organic film used as a cement to hold the quartz and solid together must provide a continuous sound transmission path. Organic materials such as stopcock grease, salol, natural rubber with Vaseline and

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** This paper represents a portion of a thesis submitted to the Rice Institute Faculty in partial fulfillment for the M.A. degree.
Table I.

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others have worked with varying success in the relatively high temperature regions. These materials may work very well even as low as 80°K, but the differential contraction usually sets in strains sufficient to cause the quartz to break away from the solid or to fracture the solid if measurements are made in a fragile material such as an alkali halide crystal. Galt encountered this difficulty in his measurements on KBr.

By using a mixture of ether, isopentane and ethyl alcohol (EPA) in amounts by volume of 5-6-2, as a cement, it has been possible to obtain measurements in beryllium as low as 3°K. The technique is to apply a liquid film of the EPA to the sample after the sample has already been cooled to about 120°K and has already undergone a considerable amount of its thermal contraction. For the ultrasonic measurements in beryllium the additional thermal contraction that takes place between 100°K, where the EPA begins to solidify, and the liquid helium range did not set in sufficient strains to break the quartz-to-solid bind. However the EPA cement is not suitable for all materials, for measurements in pure metallic tin were unsuccessful below 40°K due to the quartz breaking away from the solid at that temperature.

**VELOCITY MEASUREMENTS**

In the first longitudinal velocity measurements, an ordinary stopcock grease cement was applied as a very thin layer between the quartz crystal and the beryllium specimen. This worked very well down to about 130°K, but the thermal contraction between the quartz and the specimen caused the bind to break so that sharp echoes could not be obtained. Using the EPA mixture the longitudinal velocities could be measured from the very low temperatures to 250°K. This mixture served as a liquid transmission medium from 100°K to 250°K and as a solid cement below 100°K. There is less chance of

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* J. K. Galt, Phys. Rev. 73, 1460 (1948).

The writer wishes to express his gratitude to Dr. E. S. Lewis of the Rice Institute Chemistry Department for suggesting this material.

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Table II.

<table>
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<th>Temp. °K</th>
<th>Shear modulus $\mu \times 10^{13}$ dyne/cm$^2$</th>
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the very low temperatures, further careful measurements could be obtained during the very slow warm-up period of the cryostat. Temperatures were measured with vapor pressure and constant volume thermometers. The thermometer bulbs were placed next to the beryllium sample. The longitudinal and transverse velocities are given in Table I.

By considering the 3-in. quartz crystal as a vibrating piston in a plane wall and following the analysis due to Morse,\(^7\) the approximate intensity distributions in the longitudinal and transverse sound beams have been obtained. The computed angular width of 3.6° between the half-intensity points of the longitudinal beam accounts for pulse distortion only after the second echo. In the transverse case, a 3° beam width introduced pulse distortion after the third echo. In both cases, however, the intensities of the second echoes were very small because of effects apparently due to flaws in the beryllium sample. Because of this only a qualitative measure of the ultrasonic attenuation could be obtained. Nevertheless, any appreciable change in attenuation would manifest itself in a noticeable change in the relative amplitudes of the echoes. No such change in attenuation was observed in the region of the specific heat hump of beryllium between 10^9 K and 14^9 K.

ELASTIC MODULI

The adiabatic elastic moduli and compressibility are computed directly from the measured velocities. The longitudinal waves are propagated with a velocity \(1/\rho(1/K_0 + 4\mu/3)\)\(^1\) where \(K_0\) is the adiabatic compressibility and \(\mu\) is the shear modulus. The transverse waves are propagated with a velocity \((\mu/\rho)\)\(^1\). The length of the beryllium sample was assumed to vary with the temperature according to the linear coefficient of expansion down to 80^9 K. In the low temperature range below 80^9 K an extrapolation of the length was made assuming that the coefficient of expansion became zero at 0°K according to the Nernst thermodynamic theory. The density \(\rho\) was corrected on the basis of this extrapolation. Although some small error is introduced by this approximation, it is considerably less than the experimental error in the velocity measurements. The shear modulus and adiabatic compressibility are given in Table II. Since these quantities involve the squares of the velocities, the uncertainty in their values is at least twice as great as the velocity errors in Table I.

The Poisson's ratio was computed from

\[
\sigma = \frac{(\nu/\rho)^2 - 2}{2(\nu/\rho)^2 - 2}
\]

The value came out to be extremely small because of the relatively high transverse velocity. The value over the whole temperature range was only 0.01±1.0 percent, and the variation from 23°K to 300°K was 2 percent of the value.

CONCLUSIONS

The velocity measurements in beryllium are interesting because of the high longitudinal velocity of 12,600 meters per second. Only a few materials propagate a sound velocity higher than beryllium. The velocity in diamond has been computed from the elastic constants to be about 16,000 meters per second although we believe no direct measurements have been published.

The inaccuracies caused by pulse distortion suggest measurements at a frequency higher than 10 Mc/sec. so as to obtain a narrower sound beam and to reduce wall reflection effects that cause the pulse distortion.

The adiabatic compressibility values are accurate to 1 percent or better. Knowledge of the grain structure of the particular beryllium sample used was not available but the assumption of isotropy does not appear unreasonable in view of Roth's\(^8\) findings that there is negligible velocity dependence on the grain structure. Nevertheless the adiabatic compressibility value at 300°K in Table II is 15 percent higher than the isothermal value of 8.85×10^-12 cm^2/dyne, measured by Bridgman\(^9\) at 30°C. Bridgman's specimen may not have been as pure as the 99.2 percent purity beryllium used in the present experiments for he states that in his sample there were occlusions of the salt from which the sample had been formed. However the 15 percent difference warrants further investigation, perhaps on a different frequency and with different samples.

There was no noticeable change in the attenuation or velocity in the region of the specific heat hump between 10^9 K and 14^9 K. We may conclude that, if the specific heat hump is caused by lattice changes, then such changes must leave the velocity of sound unaffected to within the error of our measurements (1 percent). On the other hand, if the specific heat hump is caused by electronic transitions, then the attenuation of sound must be unaffected by such effects.

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