impedances. These curves make relatively simple the selection of the value of T corresponding to $n=0$. Calculations for the phase angle γ are also shown.

2.2 Merit of Arrangement for Operation with $p=1$

Although considerable simplification of the electronic equipment results from making $p>1$ (since the gate and associated dc pulse generator circuit are not needed) the advantages of operating with $p=1$ are such that the more complicated circuitry is advisable. Here every echo is used instead of every other one ($p=2$), for example. Thus, the resonance indication can be set with greater precision. Furthermore, it is possible to vary the number of echoes in the "observing" time slot by interrupting the applied pulse sequence after a predetermined number of pulses have actuated the transducer. Thus, echoes badly deteriorated as a result of spurious pulses resulting from beam spreading, for example, are eliminated. In addition (see reference 2) there is less difficulty in selecting the proper value of T for the $n=0$ condition.

2.3 Measurement of Variations of Delay and Velocity

Equation (2.2) may now be used to solve for *ratios* of delay which in turn yield ratios of wave velocity V provided the change in length l vs temperature or pressure is known. Thus,

$$V/V_0=(l/l_0)\cdot(\delta_0/\delta). \tag{2.3}$$

In the above the subscript 0 indicates the initial values.

From Eq. (2.2), ($n=0$)

$$\frac{\delta_0}{\delta}=\frac{(T_0/p)+\gamma_0/360f_0}{(T/p)+\gamma/360f}. \tag{2.4}$$

Since $\gamma/360f$ is usually quite small compared to T/p Eq. (2.4) may be written with sufficient accuracy[4] as

$$\frac{\delta_0}{\delta}=\frac{T_0}{T}\left(1+\frac{p\gamma_0}{360f_0T_0}-\frac{p\gamma}{360fT}\right). \tag{2.5}$$

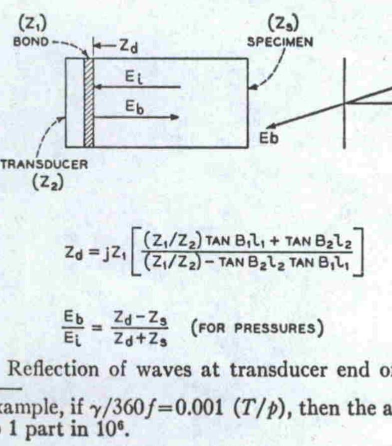

FIG. 2. Reflection of waves at transducer end of specimen.

[4] For example, if $\gamma/360f=0.001\ (T/p)$, then the approximation is good to 1 part in 10^6.

TABLE I. Values of phase angle γ for frequencies near resonance of transducer.

Conditions	γ ($f=0.99f_r$)	γ ($f=f_r$)	γ ($f=1.01f_r$)
$Z_1=2.7\times10^5$, $B_1l_1=20°$	$-3°32'$	$-7°20'$	$-11°40'$
$Z_1=5.4\times10^5$, $B_1l_1=10°$	$-3°28'$	$-7°28'$	$-10°52'$

Experimentally, one now measures T as the environment of the specimen changes. It is to be noted, however, that the pulse frequency f is a parameter that can be varied at will. Furthermore, as shown immediately following, by always matching f to the resonance frequency of the transducer, the phase angle γ is substantially constant so that the bracketed factor of Eq. (2.5) may usually be taken as unity to within a few parts in 10^5.

For a number of materials length changes with hydrostatic pressure have been directly measured, and these values may be used in Eq. (2.3). Much of this work has been done by Bridgman.[5] Reference 5 is only one of his many papers on the subject.

A method making use of the basic ultrasonic measurements to determine variations of length with pressure has been described by Cook.[6] Only cubic and hexagonal symmetries were considered.

For lower symmetries, (l/l_0) can first be obtained approximately from the zero-pressure elastic moduli; and values for all the moduli vs pressure then calculated. This simple method may be sufficient. Through perturbation a more accurate value for the variation of (l/l_0) can be determined, if needed, even though no distinction is made between adiabatic and isothermal elastic moduli for this calculation. As Cook[6] has pointed out, however, the difference between the two is quite small for many crystals, usually less than 1%, so that the net effect on velocities is quite small.

2.4 Analysis of Coupling Effect for Operation near Resonance of Transducers

Consider the reflection of waves from the specimen-bond interface as shown by Fig. 2. The impedance Z_d presented by the bond with transducer attached is given by[7]

$$Z_d=jZ_1\left(\frac{r\tan B_1l_1+\tan B_2l_2}{r-\tan B_1l_1\tan B_2l_2}\right). \tag{2.6}$$

Z_1 = characteristic impedance for bond; Z_2 = characteristic impedance for transducer; $r=Z_1/Z_2$; l_1 and l_2 are thicknesses of bond and transducer, respectively; and

[5] P. W. Bridgman, Am. J. Sci. 15, 288 (1928).
[6] R. K. Cook, J. Acoust. Soc. Am. 29, 445 (1957).
[7] H. J. McSkimin, IRE Trans. on Ultrasonics Eng., PGUE-5, (1957).

B_1 and B_2 are phase shift constants for bond and transducer, respectively.

At the resonance frequency of the transducer $\tan B_2 l_2 = 0$ (i.e., $B_2 l_2 = 180°$) so that Eq. (2.6) becomes

$$Z_d = jZ_1 \tan B_1 l_1. \tag{2.7}$$

For thin bonds, $\tan B_1 l_1$ may be replaced with $B_1 l_1 = \omega l_1 / V_1$ and $Z_1 = \rho_1 V_1$ (for 1 sq cm cross section) so that

$$Z_d = j\rho_1 l_1 \omega = jM\omega. \tag{2.8}$$

That is, the bond (if sufficiently thin) presents a mass loading to the specimen at the resonance frequency of the transducer. Since this mass is independent of temperature and pressure, the phase angle for the reflected waves (E_b of Fig. 2) will remain essentially constant. It is assumed, of course, that the impedance of the specimen varies only a few percent.

Table I shows the results of calculating the angle γ for frequencies $\pm 1\%$ from the transducer resonance, with parameters as follows:

$Z_1 = 2.7 \times 10^5$ mech. ohms/cm^2 (Nonaq stopcock grease at 20 Mc/sec and $T = 25°$C),

$Z_2 = 15.3 \times 10^5$ (X-cut quartz transducer),
$Z_s = 15.3 \times 10^5$ (X-cut quartz specimen),
$B_1 l_1 = 20°$ at Temp. $= 25°$C and pressure $= 1$ atm.

Calculations are also shown for the same combination of elements in an environment which has doubled the velocity for the bond and at least approximately doubled the impedance (since the density of commonly used bond materials remains substantially constant in comparison to velocity, the impedance $= \rho V$ is proportional to velocity). Here it is assumed that the impedances for the specimen and transducer vary only a few percent. Also, generalization of the results requires that the bond is thin.

It is seen that for the small frequency range assumed, γ is linear with frequency. Furthermore, γ remains constant with change in environment provided the frequency is maintained at a fixed *ratio* to the resonance frequency. Thus, the latter need not be known precisely for a given temperature. It is preferable, however, to maintain the frequency as close to the resonance as possible, particularly if losses in the bond are so high that the foregoing analysis may not be reliable.

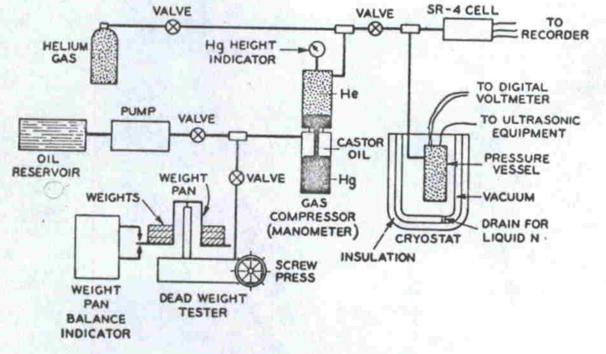

FIG. 3. Helium pressure system.

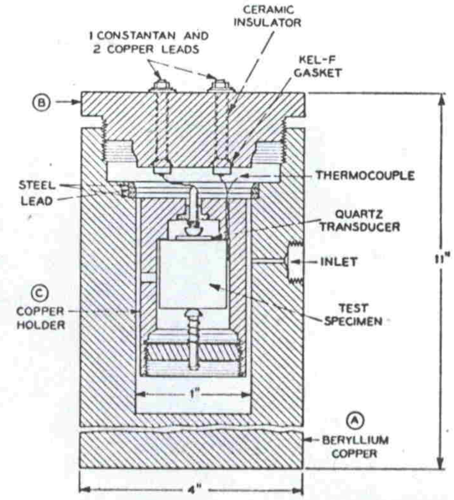

FIG. 4. Pressure vessel and specimen holder.

III. MEASUREMENTS FOR QUARTZ

3.1 Pressure System and Measurement Procedure

Pressure tests at low temperature (−195.8°C) require a pressure medium that will remain fluid throughout the range of pressures desired. Three gases which will be fluid at 30 000 psi and −195.8°C are helium, hydrogen, and neon. The last was precluded because of its cost; hydrogen, because of its explosive nature.

The helium pressure system, Fig. 3, consists of an air-driven pump, a mercury-gas compressor, a dead-weight gauge, and a pressure vessel. The dead-weight gauge measures the helium pressure to about 5 parts in 10 000. The pressure vessel, Fig. 4(a), designed for the test was made of beryllium copper "Berylco 25." It uses Bridgman-type seals[8] (lead and steel) for the closure and has one pressure inlet in the side. The closure has three cone-type electrical leads, Fig. 4(b), (two copper and one constantan) to allow a thermocouple to be placed inside the vessel.

Helium gas (tank pressure 2200 psi) was used to charge the compressor and test vessel. The tank of helium is removed and the pump forces castor oil into the compressor which then transmits the force through the mercury piston and compresses the helium. At the required pressure the air pump is disconnected and the dead-weight gauge is used to measure and maintain the pressure.

Measurements made while the temperature was changing required an accurate reading of the test speci-

[8] P. W. Bridgman, *The Physics of High Pressure* (G. Bell and Sons, London, England, 1952).

men temperature. A special copper holder, Fig. 4(c), was used which completely surrounds the test sample and assures a quick transfer of heat between the vessel and the sample. The holder was used to electrically ground the evaporated plating on the transducer face of the specimen and to hold the thermocouple in contact with the test specimen. The leads of the thermocouple pass through the pressure vessel to an ice bath and to a digital voltmeter which reads the output in μ v. Calibrating runs were made using a previously calibrated test thermocouple.

Measurements vs pressure at room temperature were obtained in an air conditioned laboratory at 25.0°±1°C. The pressure vessel, with the test specimen, was placed inside a barricade with an opening for the thermocouple and power leads. Varying the pressure (in 2000-psi steps) causes an internal temperature change in the pressure medium. After approximately 5 min, the temperature returns to within ½° of its original value (i.e., 25.0°C).[9] The basic frequency readings (cw oscillator—Fig. 1) were taken when the temperature was stable. Readings were taken for different pressures (increasing and decreasing) until all the data were obtained.

Measurements vs temperature at atmospheric pressure were obtained using the same setup, except that the pressure vessel was placed into a three-chamber cryostat, Fig. 3. The inner chamber holds the liquid nitrogen, the second chamber was held under partial vacuum, and the third chamber was filled with mica insulation. The pressure vessel was placed in the inner cryostat and the system flushed with helium gas to remove any moisture, oxygen, and nitrogen which may freeze and affect the results. Liquid nitrogen was introduced into the inner cryostat to cool the pressure vessel and the test specimen. The temperature and frequency readings were taken as the specimen was slowly cooled (keeping the pressure at atmospheric) until the inner cryostat was filled with liquid nitrogen and a stable equilibrium point (−195.8°C) was obtained. When the readings were completed, the liquid nitrogen was released into the third chamber of the cryostat and the temperature and frequency readings were recorded as the test specimen warmed up.

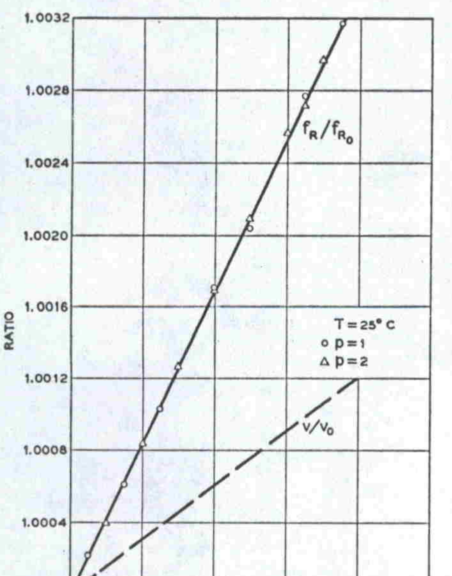

FIG. 5. Frequency and velocity ratios vs pressure for longitudinal waves propagated along the X direction of quartz, $T=25$°C.

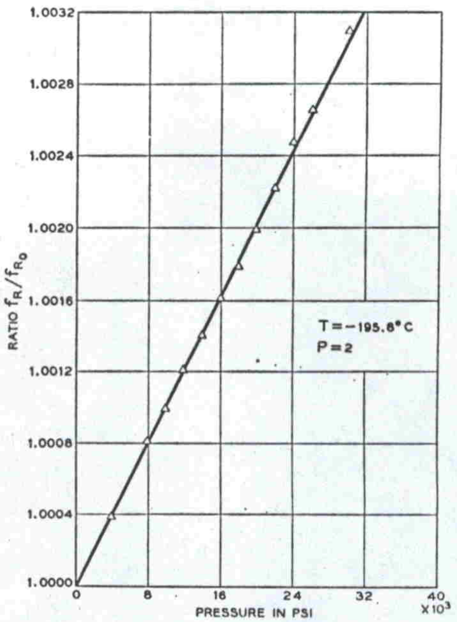

FIG. 6. Frequency ratio f_R/f_{R_0} vs pressure for longitudinal waves propagated along the X direction of quartz, $T=-195.8$°C.

Measurements were taken at liquid nitrogen temperature (−195.8°C) with pressures up to 30 000 psi, using the setup above. In order to prevent leakage the pressure vessel was charged with at least 10 000 psi before being cooled to −195.8°C. This keeps the gaskets under stress while they are contracting as the temperature is lowered. Measurements were taken after the vessel was completely immersed in liquid nitrogen and the thermocouple was at equilibrium. With each change in pressure there is a small temperature change, and one must wait a period of time for equilibrium as before. Once the gaskets have seated at −195.8°C the pressure may be raised or lowered, without any leaking, to obtain the complete data.

3.2 Results for Longitudinal Waves Propagated along *X* Direction of Quartz

For these experiments a specimen of quartz was used with dimensions 0.550, 0.560, and 0.570 in. in the X, Y, and Z directions, respectively. An X-cut 20-Mc quartz

[9] P. Andreatch and R. N. Thurston, J. Chem. Eng. Data (to be published).

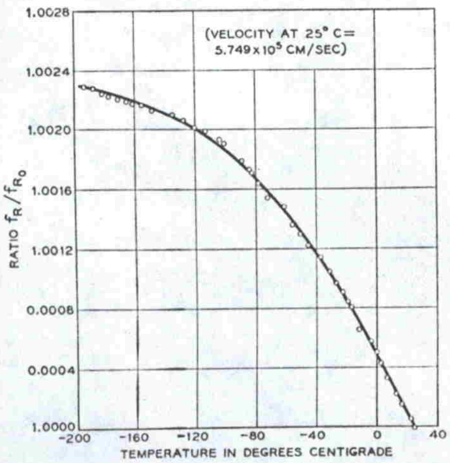

FIG. 7. Measured frequency ratio vs temperature for longitudinal waves propagated along the X direction of quartz, pressure $=1$ atm.

transducer 1 cm in diameter was attached to the specimen with Nonaq stopcock grease.

The basic quantity measured was the repetition rate frequency f_R of the pulses. This is exactly equal to $1/T$; hence, the quantity T_0/T in Eq. (2.5) may be replaced by f_R/f_{R_0}. Data for both $p=1$ and $p=2$ were obtained.

Figures 5 and 6 show plots of f_R/f_{R_0} vs pressure for 25°C and $-195.8°$ (liquid N). It is to be noted that the data for $p=2$ and $p=1$ agree very well. Because the effect of coupling to the transducer was found to be quite small it was found satisfactory to hold the wave frequency constant during the runs and to match it to the resonance frequency at 30 000 psi. The indicated correction (not shown) would decrease the slope of the ratio plot by less than $1\frac{1}{2}\%$, corresponding to an absolute change in ratio of only 4×10^{-5}.

From Bridgman's[5] measurements of length contraction vs pressure for quartz (perpendicular to the "c" axis) the value of (l/l_0) was obtained and the ratio of velocities computed [Eq. (2.3) and Fig. 5].

Figure 7 shows the basic frequency ratio vs temperature.

3.3 Results for Shear Waves Propagated along the *Y* Direction of Quartz

The same specimen was used for these measurements, but with a 20-Mc Y-cut quartz transducer transmitting

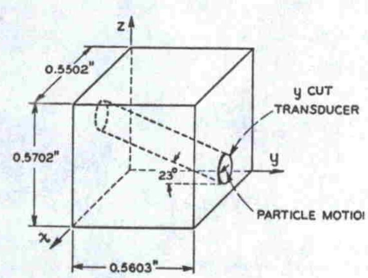

FIG. 8. Arrangement for XY shear waves.

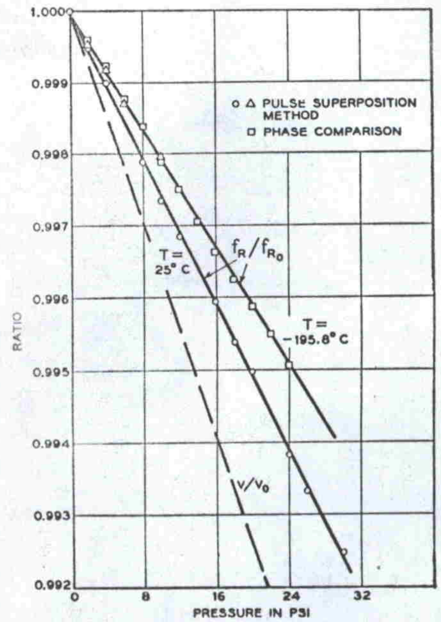

FIG. 9. Frequency ratio f_R/f_{R_0} vs pressure for XY shear waves propagated along the Y direction of quartz.

shear waves in the Y direction with particle motion along X. Since the energy flow for this case[10] is in a direction lying in the YZ plane but making a 23° angle with Y, the transducer was placed near one edge of the cube as shown by Fig. 8. This made certain that the major portion of the beam struck fully on the reflecting "Y" faces.

Figures 9 and 10 show the variation of f_R in ratio form. Here the almost negligible correction for transducer coupling would decrease the magnitude of the

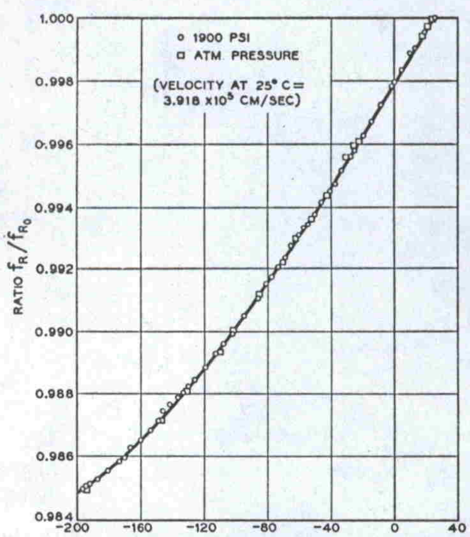

FIG. 10. Frequency ratio f_R/f_{R_0} vs temperature for XY shear waves propagated along the Y direction of quartz.

[10] For some experimental work, refer to Anderson–Shaw Labs., Interim Engineering Report on Solid Supersonic Delay Line, 1950—Armed Services Tech. Info. Agency ATI-98044.

slope by only 1%. Agreement with Susse's data is good.[11]

3.4 Resonance Frequencies of X- and Y-Cut Quartz Transducers

The data of Fig. 5 *et seq.* give also the variation of resonance frequencies vs temperature and pressure for X- and Y-cut quartz transducers; however, there may be a very slight difference if the driver electrical impedance (measured at the transducer terminals) is too low. This difference arises from the fact that the velocity of waves in the comparatively large specimen depends on the piezoelectric coupling in a slightly different way than for the transducer with electrodes shorted or

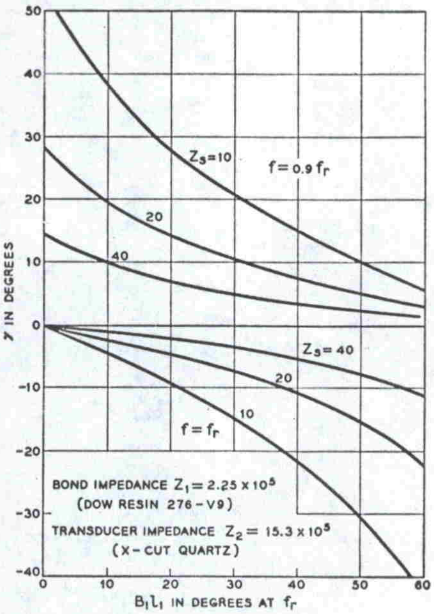

FIG. 11. Phase angle γ vs thickness of bond for longitudinal waves.

terminated in a low impedance. This can be seen from the equivalent circuit of a thickness-type transducer as described by W. P. Mason.[12] For the purpose of velocity measurements as described in this paper, however, the data presented are more than adequate.

Work is being continued which will result in a complete set of velocity measurements vs temperature and pressure. These data will make possible the determination of all elastic moduli for quartz within the ranges 25° and −195.8°C, and 0 to 30 000 psi.

APPENDIX A

Calculations for ΔT vs Thickness of Bond

For determining the value of T corresponding to $n=0$ [see Eq. (2.1)] one compares experimental and theoretical values of ΔT as the wave frequency is varied.

[11] C. Susse, J. phys. radium **16**, 348 (1955).

[12] W. P. Mason, *Electromechanical Transducers and Wave Filters* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1948), 2nd ed.

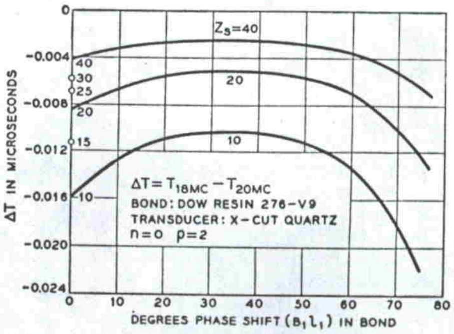

FIG. 12. ΔT vs $B_l l_l$ for longitudinal waves.

From theory,[2] the change in T caused by a change in frequency from f_H to f_L is

$$\Delta T=\frac{1}{f_L}\left(n-\frac{p\gamma_L}{360}\right)-\frac{1}{f_H}\left(n-\frac{p\gamma_H}{360}\right). \qquad \text{(A1)}$$

Letting $f_H=f_r$, the transducer resonance frequency, and $f_L=0.9f_r$, γ_L, and γ_H can be evaluated[7] and values of ΔT obtained for $n=0$, using Eq. (A1).

Figures 11–14 show the results obtained for longitudinal and shear waves, it being assumed that X-cut and Y-cut quartz transducers, respectively, are used. (Similar calculations could be made for other transducers.) All calculations are for a bond made of Dow resin 276-V9, a viscous polystyrene fluid which serves quite well for both types of waves at room temperature and pressure.

Figures 12 and 14 are for $p=2$. For $p=1$, values of ΔT are exactly one-half those shown.

By way of illustration, measurements of ΔT for shear waves were made for a germanium specimen approxi-

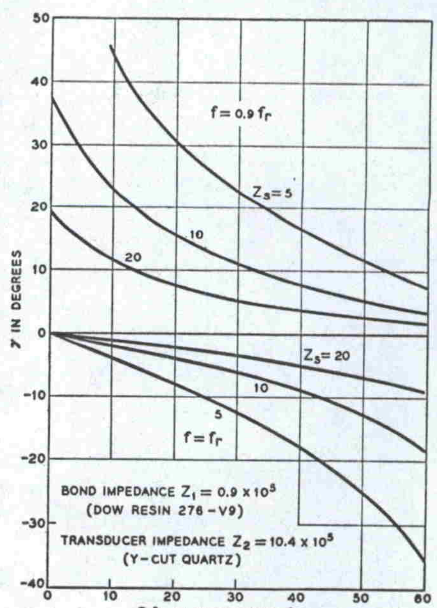

FIG. 13. Phase angle γ vs thickness of bond for shear waves.

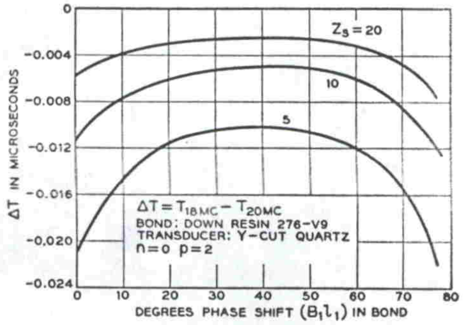

FIG. 14. ΔT vs B_1l_1 for shear waves.

mately a cube ½ in. on a side. Propagation along the [100] direction was used with an impedance Z_s of 18.8×10^5 mech. ohms/cm². From Fig. 14 it is seen that the measured -0.003 μsec value of ΔT corresponds to $n=0$ with an indicated bond thickness of 20° (complete plot of data shown by Fig. 15). This thickness can only be approximate since the curve of ΔT vs Bl is so flat; furthermore, departures from the ideal would cause uncertainties. This point must be *stressed* inasmuch as excessive loss in the bond can sometimes lead to choosing the wrong repetition rate frequency for $n=0$. A very thin bond is helpful. With the Dow resin 276-V9 this can be obtained by using a polished transducer and by heating the bond to about 60°C under moderate pressure to squeeze out excess material. For actual pressure and temperature runs, a bond suitable for the range of parameters would, of course, be used.

The value of γ for a 20° bond is seen to be $-2°$ (Fig. 13) assuming operation precisely at the 20-Mc resonance frequency of the transducer. The round-trip delay δ [Eq. (2.2)] is then

$$\delta = (14.6757\times10^{-6}/2) - [2/360(20\times10^6)],$$

or

$$\delta = (7.33783 - 0.0002778)\times10^{-6} = 7.3376\ \mu\text{sec}.$$

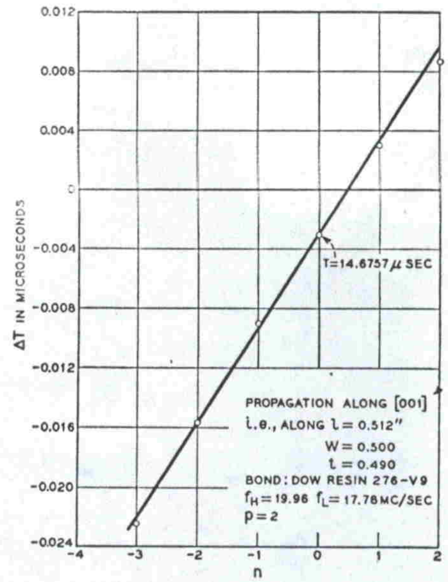

FIG. 15. Measured values of ΔT vs n for shear waves in a germanium specimen.

This leads to a velocity of 3.543×10^5 cm/sec based on a thickness of 0.5118 in. ±0.05%, and neglecting diffraction effects which should be only a few parts in 10^4.[13]

[13] H. J. McSkimin, J. Acoust. Soc. Am. **32**, 1401 (1960).