A New Absolute High Pressure Gauge

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ABSTRACT

This is a preliminary description of a new absolute pressure measurement system which is based on the simultaneous measurement of length and ultrasonic transit times. The system has been used to determine the transition pressure of pure mercury at 0°C at 7571.2±1.6 bars. It is particularly advantageous to use as a device for calibrating secondary gauges such as manganin gauges inasmuch as it gives a highly accurate measurement of nonlinearities in such secondary gauges. The gauge calibrated in the present study reaches a maximum deviation from linearity of 11.6 bars at a pressure midway between atmospheric pressure and the 0°C mercury melting pressure.

I. INTRODUCTION

The basis of this pressure measurement system is the equation which defines isothermal bulk modulus $B^T(P)$ according to

$$dP = -B^T(P) \frac{dV}{V} \quad (1)$$

If the material studied is elastically isotropic or

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cubic, then the linear compressibility is isotropic (so that $3 d\ell/\ell = dV/V$) and

$$dP = -3 B^T \frac{d\ell}{\ell} \quad (2)$$

The adiabatic bulk modulus can be measured ultrasonically by measuring simultaneously transit times and transit lengths.

The adiabatic bulk modulus for a cubic crystal is

$$B^S = C_\ell - 4C_s/3 = \rho \ell^2 \left(\frac{1}{\tau_\ell^2} - \frac{4}{3\tau_s^2} \right) \quad (3)$$

Here $C_\ell = (C_{11} + 2C_{12} + 4C_{44})/3$ and $C_s = C_{44}$. Moreover, ρ is the density and ℓ the transit length while τ_ℓ is the transit time for the [111] longitudinal wave and τ_s is the transit time for the [001] shear wave.

In actual experiments, specimen lengths are different, but for convenience of presenting the mathematical expressions, we write them as if the lengths are the same.

The isothermal bulk modulus is

$$B^T = \frac{B^S}{1+\Delta} \quad (4)$$

where

$$\Delta = \frac{\beta^2 B^S T}{\rho C_p} \quad (5)$$

Here β is the volume thermal expansion coefficient, T is the absolute temperature and C_p is the specific heat at constant pressure. For silicon Δ is less than 0.0011 on the pressure range $0 \leq P \leq 8$ kbar. Moreover, as shown in the theoretical section $\Delta(P)$ can be readily calculated.

Thus, by combining Equations (2)-(4) and integrating, we

obtain the pressure in the form

$$P = P_1 - 3\rho_1 \ell_1^3 \int_{\ell_1}^{\ell} \left(\frac{1}{\tau_{\ell}^2} - \frac{4}{3\tau_s^2} \right) \left(\frac{1}{1+\Delta} \right) \frac{d\ell}{\ell^2} . \quad (6)$$

Here ρ_1 and ℓ_1 denote the density and length at atmospheric pressure P_1 . By increasing the pressure in small (but unknown increments) and simultaneously measuring τ_{ℓ} , τ_s and ℓ at the same (unknown) pressure, and then integrating, the pressure P is obtained.

Historical Aspects

The idea for the absolute pressure gauge occurred to us in 1966. The background is of interest. We were at that time attempting to set up an analytical procedure for obtaining B_0^S (the second pressure derivative of the bulk modulus evaluated at zero pressure) for cubic crystals from ultrasonic transit measurements which were being made in our laboratory. To get B^S at pressure requires that the length of the specimen (and its density, which can be computed from its density at atmospheric pressure if the length is known) be known at pressure as is clear from Equation (3). Prior to this time the method used was to compute ℓ from the measured value of ℓ_1 and of the bulk modulus B_1 (at atmospheric pressure)¹ using the relationship

$$\ell = \ell_1 \left[1 - \frac{(P-1)}{3B_1^T} \right] . \quad (7)$$

We call this the boot strap method.

Since one result of this approach was to measure the first pressure derivative, a clearly better approach was to assume

$$B^T = B_0^T + B_0^{T'} P \quad (8)$$

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and to use this with the first approximate value of $B_0^{S'}$ from Equation (7) in Equation (1) to obtain an expression for $V/V_1 = (\ell/\ell_1)^3$ which could then be used iteratively to get $B_0^{S'}$.² [The quantity actually measured is B_0^U . This is the isothermal pressure derivative of the adiabatic bulk modulus evaluated at zero pressure. It can be converted to $B_0^{T'}$ (the isothermal pressure derivative of the isothermal bulk modulus) or to $B_0^{S'}$ (the adiabatic pressure derivative of the adiabatic bulk modulus).] However, we realized that this extended boot strap approach was not the final solution. At this point (late May of 1966) the senior author (ALR) journeyed to Albuquerque to carry on some research at the Sandia Corporation on equation of state. The purpose of this work was to compare the equation of state obtained from ultrasonic measurements to the equation of state obtained from shock measurements at ultra high pressures.^{3,4} In the process the senior author became conditioned to writing Hooke's law for the case of an applied hydrostatic pressure in the form of Equation (1). Upon returning to Cornell in June 1966 he found that his student (RCL) and Research Associate Paul S. Ho had made considerable progress toward analyzing ultrasonic transit time data. The latter had found that Cook⁵ had already developed a scheme based on the inverted form of Equation (2) but that this apparently had not been used. The former meanwhile independently developed an iterative scheme based on an equation of state which was based on $B = B_0^S + B_0^{S'} P + B_0^{S''} P^2/2$. However, he did not believe that convergence could be guaranteed by this further extended boot strap approach and he suggested that what we should really do is to measure the length simultaneously with the transit

times. We decided jointly that we should proceed to see how well this could be done. It was our decision to see if we could design a system in which length changes could be measured to 2 parts in 10^8 even when the displacement was large.

Shortly thereafter it occurred to us that if length were measured at an unknown pressure at the same time that the transit times were measured, this could serve as an absolute pressure gauge according to Equation (6). Soon after we also noted that the simultaneous measurement of ultrasonic transit times as a function of pressure and temperature provided a direct measurement of Gruneisen's parameter as a function of pressure. We now had considerable more motivation to design and construct such a system.

The general design of the system was completed in the fall of 1966. We were very fortunate to have Professor Ralph Simmons visit us, and discuss and criticize the system design at that time. Moreover, our former colleague Dr. P. B. Ghate who was working at Texas Instruments assured us that the necessary meter long silicon crystals could be grown. We then proceeded to construction and, after some considerable delays (caused primarily by the need to control temperature of a pressure vessel containing a meter long specimen to about one millidegree Kelvin) achieved nearly our original design requirement.

II. THEORY

Basic Concepts

In principle any solid with an isotropic linear compressibility (which behaved reversibly under application of pressure) could be used for the working media of the absolute pressure gauge,

although it will become clear later why a material should be chosen with Δ as small as possible.

The quantity Δ is exactly given by

$$\Delta = \frac{\beta^2 B^S T}{\rho C_P} \quad (9)$$

Use is made of the exact thermodynamic relations⁶

$$\left(\frac{\partial \beta}{\partial P}\right)_T = -\left[\frac{\partial}{\partial T}\left(\frac{1}{B}\right)\right]_P \quad (10)$$

and

$$\left(\frac{\partial C_P}{\partial P}\right)_T = -T\left[\frac{\partial^2}{\partial T^2}\left(\frac{1}{\rho}\right)\right]_P = -T\left[\frac{(\partial \beta / \partial T) + \beta^2}{\rho}\right]_P \quad (11)$$

to evaluate $\beta(P)$ and $C_P(P)$. In our analysis it is the quantity Δ as a function of ℓ which must be known to carry out the integration in Equation (6). We therefore replace Equations (10) and (11) by

$$\left(\frac{\partial \beta}{\partial \ell}\right)_T = -\left[\frac{3}{\ell B T} \frac{\partial B}{\partial T}\right]_P \quad (12)$$

and

$$\left(\frac{\partial C_P}{\partial \ell}\right)_T = 3T B_T \left[\frac{(\partial \beta / \partial T) + \beta^2}{\rho \ell}\right]_P \quad (13)$$

We also note that B^S/ρ can be replaced by

$$B^S/\rho = \ell^2 \left(\frac{1}{\tau_\ell^2} - \frac{4}{3\tau_s^2}\right) \quad (14)$$

Two approaches can be used to evaluate $\Delta(\ell)$.

Linear Approach

Here we let

$$\Delta = \Delta_1 + \left(\frac{\partial \Delta}{\partial \ell}\right)_{\ell_1} (\ell - \ell_1) \quad (15)$$

The evaluation of Δ_1 requires that T , ρ_1 , B_1^S and also β_1 and C_{p_1} be known. The evaluation of $(\partial\Delta/\partial\ell)_{\ell_1}$ requires that the length derivatives $[\partial(B^S/\rho)/\partial\ell]_{\ell_1}$, $(\partial\beta/\partial\ell)_{\ell_1}$ and $(\partial C_p/\partial\ell)_{\ell_1}$ be determined also. Since the first of these is directly measured in our experiment [see Equation (14)], its length derivative is easy to obtain. The other two pressure derivatives can be obtained from measurements made at pressure P_1 (atmospheric pressure) using Equations (12) and (13). This requires in addition a knowledge of the temperature dependence of the quantities β , B^S/ρ and C_p at the pressure P_1 , i.e., when the length equals ℓ_1 .

General Approach

In the general approach, all of the quantities in Equation (9), or their thermodynamic equivalent, are measured directly as a function of length. No linear assumption such as used in Equation (15) is made. The quantity B^S/ρ is measured directly as a function of length at a given temperature [see Equation (14)] and need not concern us further.

Next, we utilize the fact that length and transit times can be measured at both different pressures and different temperatures. From such measurements β and $\partial\beta/\partial T$ evaluated at a given T and ℓ can be obtained. Of course ρ can also be obtained once ρ_1 is determined. Therefore, the derivatives in Equation (12) can be obtained and from this we have

$$C_p(\ell) = C_p(\ell_1) + \int_{\ell_1}^{\ell} \left(\frac{\partial C_p}{\partial \ell} \right)_T d\ell \quad (16)$$

III. EXPERIMENT

Silicon was chosen:

(1) because the neglect of the Δ term would cause an error of less than 6 bars at 7500 bars. However, we evaluate Δ so this error is not introduced,

(2) because excellent single crystals of silicon can be grown and hence the material can be expected to behave reversibly. It might be added that the crystal used for length measurements was over one meter long.

Length Measurement

We begin with an initial discussion of the method used to obtain changes in length of such a specimen. The method is only outlined here but a comprehensive discussion is given elsewhere.⁷

Nonmagnetic pressure tubing extends from the ends of the pressure vessel and the specimen is shaped so as to move freely in the tubing (Figure 1). Special H-shaped soft magnetic cores (see Figure 2) are placed over each end and are slightly spring loaded. The stiffness constants of the springs and their initial compression is known and corrections are made for length changes caused by the spring, both at P_1 and at P . The only correction needed for the magnetic core (which changes in length as pressure is changed) is for the webbed portion of the core which is 0.01 cm thick. External to the pressure tubing are transformers (see Figure 2) which accurately locate the position of the cores by a null technique. Each transformer is mounted on a carriage (see Figure 3) which moves on a track. The carriage is displaced by an electrically driven coarse micrometer, a fine micrometer and finally a piezoelectric device. Motion as small as $20\text{\AA}$ can be produced. There are also mirrors contained on the central axes of these car-

riages and the distance between these is measured with a Fabry-Perot interferometer. The laser light beam travels in a vacuum in a highly flexible bellows. The pressure in the bellows is monitored and corrections are made for the small wavelength changes due to refractive index changes.

A reversible fringe counter is employed in a phase sensitive fashion so that fractional fringes can be measured. Figure 4 shows the fringe circles produced by modulation and the fringe spot which measures a fractional fringe. One circle is half a wavelength ($\lambda = 6329.9147\text{\AA}$). In the lower right photograph we see the fringe trace which indicates the fractional fringe count to be added to the reversible counter. We note that it has a width 4° (due to electrical and mechanical noise); this noise corresponds to a length change of less than $40\text{\AA}$. Other factors are present which increase the absolute error of length measurement so that the overall fractional resolution of the system on the meter long specimen is 4×10^{-8} . The entire pressure vessel is enclosed in a constant temperature bath controlled to $2 \times 10^{-3}^\circ\text{C}$. It should be noted that this system has the capability of such fractional length resolution even if the specimen length changes by 4 cm. The overall system (without the insulating cover) is shown in Figure 5. The entire framework is mechanically isolated from the room.

The length ℓ_1 at atmospheric pressure is measured interferometrically. The fractional error of the original length of the specimen, ℓ_1 , is 1×10^{-5} .

Transit Time Measurements

The silicon specimens for the ultrasonic measurements are

made from silicon crystals of the same purity as that used in the length measurement. Four crystals were sliced from the single crystal. Each of these crystals are oriented first by the Laue technique and then by the single spot diffraction technique to within 0.1° of the desired orientation. Two crystals with (100) faces and two with (111) faces were prepared. The surfaces were lapped parallel to within $0.2'$. The distance between these surfaces is measured by a Van Keuren light-wave micrometer to 2×10^{-5} cm. The transducers were bonded with glycerine for the longitudinal wave and salol (phenyl salicylate) for the shear wave. The four specimens were placed in a four crystal jig⁸ which was then placed in the pressure vessel. In this way both τ_s and τ_l were measured in duplicate at the same pressure.

The pulse superposition method⁹⁻¹¹ was used to measure the transit times. The fractional transit times in a composite (quartz-bond-silicon) could be easily measured to better than 10^{-5} , but the reproducibility was within 2×10^{-5} when bonds were reformed and a new composite was made. The reproducibility is the important quantity to consider when an absolute pressure (such as the mercury point at 0°C) is measured. Possible sources of absolute errors in τ are discussed elsewhere.¹²

However, (and this must be emphasized), the fractional error in change in transit time as a result of applying pressure is considerably less than would be the case if each measurement at pressure were known to a part in one hundred thousand. Let us therefore consider the representation of the pressure resistance of the manganin gauge in the form

$$P = a\left(\frac{\Delta R}{R_1}\right) + b\left(\frac{\Delta R}{R_1}\right)^2 \quad (17)$$

See Figure 6.

For purposes of discussion let us assume that the manganin point is fixed exactly (by accident, for instance, since all measurements have some possible random and absolute errors). Let us compare the relative reliability of the nonlinear coefficient b measured by our pressure gauge and that measured by the free piston gauge. Another convenient measure of the nonlinearity is the quantity ΔP_M shown in Figure 6. This is the maximum deviation of the guage from linearity for pressures between $P = P_1$ and $P = P_{Hg}$. It can be shown¹³ that

$$\Delta P_M = \frac{-b(P_{Hg} - P_1)^2}{4a_o^2} \quad (18)$$

where

$$a_o = \frac{a + \sqrt{a^2 + 4b(P_{Hg} - P_1)}}{2} \quad (19)$$

Let us assume a random error of 2 bars at each pressure for the free piston gauge. Thus the value of ΔP_M would be known to at best ± 1 bar. The random error of an individual reading in the present system is 0.04 bars or less. Consequently, ΔP_M can easily be measured to 0.05 bars. It is important to know ΔP_M accurately in many applications. For example, if the pressure derivative of the bulk modulus of iron is measured by length change, an error of 2 bars in ΔP_M would give an error of nearly 10% in B'_0 .¹⁴

Manganin Pressure Sensor

In order to achieve high sensitivity a bridge consisting of four manganin arms was created. This system is described in de-

tail elsewhere.¹³ It has a sensitivity of better than 0.04 bars. The manganin wire in the two coils within the pressure vessel is passed directly through cone seals¹⁵ and connected to the outer two arms outside the vessel. All four arms are placed in a temperature controlled environment held to within $\pm 0.01^\circ\text{C}$.

Mercury and Mercury Cell Calibration

The mercury used was triply distilled and 99.999% pure.

The mercury pressure cell was enclosed in a pressure vessel with a 2.54 cm internal diameter. The mercury container was a U shape glass capillary tube 30 cm long with a 0.06 cm internal diameter.

The pressure vessel was placed in a plastic ice chamber 34 cm in diameter and 56 cm high (the inner chamber). The ice was made from distilled water which had previously been deionized. This was surrounded by the outer ice chamber which had an internal diameter of 52 cm and was 100 cm high. The outer chamber in turn was surrounded by a styrofoam box of dimensions 80x80x125 cm.

After an initial experiment, the mercury point was determined to within a few bars. The system was then raised to just above the mercury point. Then the pressure was decreased at a rate of a small fraction of a bar per hour. At the mercury point there was a pressure arrest (zero rate change of pressure versus time) with a duration of one hour or more. Even at this slow rate of transformation there was a small decrease in temperature due to absorption of heat and this was estimated to cause a pressure error of 0.3 bars which was added to the value obtained. In order to minimize temperature correction, long waiting periods and an essen-

tially leak-proof pressure system was a necessity.

IV. DATA ANALYSIS

Evaluation of Δ_1

The expression for Δ is given in Equation (9). The volume thermal expansion coefficient for silicon used here is the value given by Batchelder and Simmons;¹⁶ other values are given by Gibbons¹⁷ and Ibach.¹⁸ The adiabatic bulk modulus at atmospheric pressure was measured by the present authors. The value used for the density of silicon is that of Henins¹⁹ corrected to our temperature. At 25°C and one atmosphere he gives a value of $\rho = 2.329002 \text{ g/cm}^3$. [Values extremely close to this at 25°C are given by Smakula and Sils²⁰ ($\rho = 2.32902 \text{ g/cm}^3$), Straumanis et al²¹ ($\rho = 2.3289 \text{ g/cm}^3$), and Stebler²² ($\rho = 2.32881 \text{ g/cm}^3$).] Although it is not necessary to have this accuracy for ρ to evaluate Δ_1 , it is necessary to know ρ extremely accurately when evaluating P using Equation (6). The specific heat value is extrapolated from the data given by Flubacher, Leadbetter and Morrison.²³

The value of Δ_1 at 31.7°C obtained is $1.1376 \times 10^{-3} \pm 5.8 \times 10^{-6}$.

Evaluation of $(\partial \Delta / \partial \ell)_{\ell_1}$

The isothermal derivative is given by

$$\left(\frac{\partial \Delta}{\partial \ell}\right)_{\ell_1} = \Delta_1 \left[\frac{2}{\beta} \left(\frac{\partial \beta}{\partial \ell}\right) - \frac{1}{C_p} \left(\frac{\partial C_p}{\partial \ell}\right) + \frac{1}{Z} \frac{dZ}{d\ell} \right]_{\ell_1} \quad (20)$$

where

$$Z = B^S / \rho \quad (21)$$

The quantity Z is measured directly as a function ℓ [see Equation (14)] so that $\left(\frac{1}{Z} \frac{dZ}{d\ell}\right)_{\ell_1}$ is directly obtained from our measurements.

The quantity $(\frac{\partial \beta}{\partial \ell})_{\ell_1}$ can be evaluated if the length, density and transit times are known as functions of temperature [see Equation (12)] at atmospheric pressure. The data of Batchelder and Simmons¹⁶ on lattice parameter versus temperature was used for length and density variations.

The quantity $(\frac{\partial C_p}{\partial \ell})_{\ell_1}$ at the fixed temperature T can be evaluated by combining Equations (13) and (16). This necessitates knowing C_p , ρ and ℓ at atmospheric pressure and knowing $\ell(T)$ with sufficient accuracy so that $\partial^2 \ell / \partial T^2$ is obtainable. The data of Batchelder and Simmons¹⁶ was used for the latter. In this way we obtain

$$(\frac{\partial \Delta}{\partial \ell})_{\ell_1} = 6.345 \times 10^{-4} \pm 3.2 \times 10^{-6}.$$

The error in Δ_1 and $(\frac{\partial \Delta}{\partial \ell})_{\ell_1}$ can cause an error in the mercury point determination of no larger than ± 0.05 bars.

Evaluation of $P(\ell)$

The absolute pressure P is evaluated using Equation (6). Of course ℓ is varied discretely instead of continuously so that the integration is necessarily performed in some approximate way (although the accuracy may be very high). In the present case Simpson's rule was used. In this way a set of P vs ℓ or ℓ vs P data is obtained. (It should be noted that the resistance ratio of the managanin pressure sensor is measured simultaneously with ℓ so that a calibration of this managanin gauge is also obtained.)

The ℓ vs P data is then fitted to an equation of state using a special nonlinear least squares regression analysis so that now a continuous function $\ell(P)$ is obtained.

In the present case we used Murnaghan's equation which is a two parameter equation of state based on the assumption that the isothermal bulk modulus varies linearly with pressure:

$$B^T = B_0^T + B_0^{T'} P \quad (22)$$

The resultant equation of state obtained by combining Equations (1) and (22) is

$$\frac{\rho_1}{\rho} = \left[\frac{B_0^T + B_0^{T'} P}{B_0^T + B_0^{T'} P_1} \right]^{1/3 B_0^{T'}} \quad (23)$$

The fact that Murnaghan's equation is probably the best two parameter fit known is described elsewhere by Anderson²⁴ and Ruoff.^{3,4}

Three Parameter Fit of $\rho(P)$

We have in a somewhat different manner fitted $\rho(P)$ to an equation of state based on

$$B^T = B_0^T + B_0^{T'} P + B_0^{T''} \frac{P^2}{2} \quad (24)$$

as Ruoff⁴ had considered earlier [see his Equations (10)-(14)] for fitting shock data [except here we are concerned with pressure being varied isothermally (rather than adiabatically)].

As soon as the set of P vs ρ data is obtained we compute from our measured values a set of B^S vs P data inasmuch as B^S is given by Equation (3). Then, using Equations (15) and (4) we can convert the B^S vs P data to a set of B^T vs P data. This is then fitted using a least squares analysis to Equation (24). It is also fitted using Equation (24) for the case where $B_0^{T''} \equiv 0$; this provides a check on the nonlinear least squares analysis discussed in the previous subsection.

The 0°C mercury freezing point was determined to be 7571.2 bars ± 0.8 bars (2σ random deviation). The major source of this random error is attributed to the "sluggishness" of the phase transformation. Error analysis shows an additional possible absolute error of 0.8 bars, primarily due to possible errors in measuring the absolute transit times in the crystal. Thus the final value presented here is 7571.2 ± 1.6 bars.

As a result of the nonlinear least squares fit we obtained for silicon the following rounded off values and standard deviations

$$B_0^T = 977.87 \pm 0.02 \quad (\text{at } 31.7^\circ\text{C})$$

$$B_0^{T'} = 4.04 \pm 0.01 \quad (\text{at } 31.7^\circ\text{C}).$$

As a result of converting P vs λ data points to B^T vs P data points and then fitting to Equation (24) we obtain

$$B_0^T = 978.24 \pm 0.05 \text{ kbars} \quad (\text{at } 31.7^\circ\text{C})$$

$$B_0^{T'} = 3.81 \pm 0.04 \quad (\text{at } 31.7^\circ\text{C})$$

$$B_0^{T''} = 0.06 \pm 0.01 \text{ kbars}^{-1} \quad (\text{at } 31.7^\circ\text{C}).$$

If in this analysis we had fitted to Equation (24) with $B_0^{T''} \equiv 0$, we obtain results identical to the first set given for B_0^T and $B_0^{T'}$.

Once the set of data for P, $\frac{\Delta R}{R_0}$, λ is obtained then the coefficients a and b in Equation (17) can be obtained and the maximum deviation of the manganin gauge (under calibration) from nonlinearity for pressures between $P = 0$ and $P = P_{Hg}$ can be obtained. Using Equations (18) and (19) we obtain

$$\Delta P_M = -11.6 \pm 0.1 \text{ bars.}$$

VI. COMPARISON OF RESULTS, DISCUSSION AND CONCLUSIONS

Table 1 gives values of the 0°C mercury point transition pressure.

Table 1. The 0°C Mercury Freezing Pressure

<u>Pressure (bars)</u>	<u>Source</u>
7565.4 $\pm$ 3.7	Newhall, Abbot and Dunn ²⁵
7569.2 $\pm$ 1.2	Dadson and Greig ²⁵
7571.0 $\pm$ 1.2	Yasunami ²⁷
7571.2 $\pm$ 1.6	Present

It should be noted that the Committee on Fixed Points Near Room Temperature at the Symposium on Accurate Characterization of the High - Pressure Environment held at the National Bureau of Standards, Gaithersburg, Md. in 1968 designated 7569 $\pm$ 2 bars as the accepted value for the 0°C mercury freezing pressure.

Table 2 gives values of the bulk modulus and its pressure derivative at zero pressure.

Table 2. Values of B_0^T and $B_0^{T'}$

<u>B_0^T (kbars)</u>	<u>$B_0^{T'}$</u>	<u>Source</u>
* 976.79	** 4.24	McSkimin and Andreatch ³⁰
977.87	4.04	Present Work

*Converted from B^S . Corrected for p_1 .

**Converted from $(\frac{dB^S}{dP})_T$. Corrected for p_1 . Value computed from data at atmospheric pressure and one pressure only (near 2 kbars).

Direct comparison of ΔP_M may not be meaningful although values are given elsewhere¹³ and repeated in Table 3 since it is conceivable that ΔP_M varies with the manganin sample. It is, however, also

Table 3. Values of ΔP_M

ΔP_M (bars)	Source
-10	Harwood ¹⁹
-11	NBS ³²
-21	Zeto and Vanfleet ³³
-21	Boren, Babb and Scott ³⁴
-11.6	Present Work

conceivable that ΔP_M is not very strongly dependent on the sample (assuming its purity and fabrication are nearly the same) as Bridgman proposed but rather dependent on the inaccurate measurement of pressure. At this point we do not believe the answer is known.

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REFERENCES

1. W. B. Daniels and C. S. Smith, in Physics and Chemistry of High Pressures, Gordon and Breach, Science Publishers Inc., New York (1963) p. 50.
2. A. L. Ruoff, Sandia Corp. Report, SC-RR-66-676 (June 1966).
3. A. L. Ruoff, J. Phys. Chem. Solids 28, 453 (1967).
4. A. L. Ruoff, J. Appl. Phys. 38, 4976 (1967).
5. R. K. Cook, J. Acous. Soc. Am. 29, 445 (1957).
6. W. C. Overton, Jr., J. Chem. Phys. 37, 116 (1962).
7. R. C. Lincoln and A. L. Ruoff, Rev. Sci. Instr. (Submitted for publication).
8. Y. C. Chen, R. H. Martinson and A. L. Ruoff, Rev. Sci. Instr. (to be published Nov. 1972).
9. J. Williams and J. Lamb, J. Acous. Soc. Am. 30, 308 (1958).
10. A. D. Colvin, M. S. Thesis, Rensselaer Polytechnic Institute, Troy, New York (1959).
11. K. M. Koliwad, P. B. Ghate and A. L. Ruoff, Phys. Stat. Sol. 21, 507 (1967).
12. H. J. McSkimin and P. Andreatch, Jr., J. Appl. Phys. 35, 2161 (1964); and H. J. McSkimin and P. Andreatch, Jr., J. Acous. Soc. Am. 34, 609 (1962).
13. R. C. Lincoln, Ph.D. Thesis, Cornell University, Ithaca, New York (1971).
14. R. C. Lincoln and A. L. Ruoff (In preparation).
15. R. E. Terry and A. L. Ruoff, Rev. of Sci. Instr. 43, 1379 (1972).
16. D. N. Batchelder and R. O. Simmons, J. Chem. Phys. 41, 2324 (1964).
17. D. F. Gibbons, Phys. Rev. 112, 136 (1958).
18. H. Ibach, Phys. Stat. Sol. 31, 625 (1969).
19. I. Henins, J. Res. Nat. Bur. Std. 68A, 529 (1964).
20. A. Smakula and V. Sils, Phys. Rev. 99, 1744 (1955).

21. M. E. Straumanis, P. Borgeaud and W. J. James, J. Appl. Phys. 32, 1382 (1961).
22. B. Stebler, Arkiv für Fysik 30, 91 (1965).
23. P. Flubacher, A. J. Leadbetter and J. A. Morrison, Phil. Mag. 4, 273 (1959).
24. O. L. Anderson, J. Phys. Chem. Solids 27, 547 (1966).
25. D. H. Newhall, L. H. Abbot and R. A. Dunn, in High Pressure Measurement, ed. by A. A. Giardini and E. C. Lloyd, Butterworths, Washington (1963) p. 339.
26. R. S. Dadson and R.G.P. Greig, Brit. J. Appl. Phys. 16, 1711 (1965).
27. K. Yasunami, Proc. Jap. Acad. 43, 310 (1967).
28. Nat. Bur. Std., Spec. Pub. 326 (1971) p. 313.
29. E. C. Lloyd and C. W. Beckett, Science 164, 860 (1969).
30. H. J. McSkimin and P. Andreatch, Jr., J. Appl. Phys. 35, 2161 (1964).
31. D. H. Newhall, Harwood Engineering, Private communication.
32. D. P. Johnson, National Bureau of Standards, Private communication with R. C. Lincoln.
33. R. J. Zeto and H. B. Vanfleet, J. Appl. Phys. 40, 2227 (1969).
34. M. D. Boren, S. E. Babb, Jr. and G. J. Scott, Rev. Sci. Instr. 36, 1456 (1965).

FIGURE CAPTIONS

Figure 1: Magnetic core.

Figure 2: LVDT and pressure tube assembly.

- 1) Specimen.
- 2) Ball bushing support for specimen.
- 3) Portion of specimen supported by ball bushing.
- 4) Bridgman seal-mushroom.
- 5) Bridgman seal - Indium ring.
- 6) Bridgman seal - Miter ring (one of four).
- 7) Bridgman seal - Thrust collar.
- 8) Bridgman seal - Thrust nut.
- 9) Bridgman stem - (12.7 mm. O.D. by 3.2 mm. I.D. - 316 stainless steel high pressure tubing).
- 10) Bridgman seal - Extractor clamp.
- 11) Linear variable differential transformer (LVDT).
- 12) LVDT positioning ball (for λ_0 measurement).
- 13) LVDT core ("H" shape core).
- 14) LVDT core retaining spring.
- 15) Gland nut for connector.
- 16) Tuning collar for connector.
- 17) Tubing connector.
- 18) End plug.

Figure 3: LVDT and laser mirror carriage.

Figure 4: Fringe traces. The position of the "spot" in the lower two figures indicates the fraction of one fringe. (Movement of the spot through 360° is one fringe.)

Figure 5: Open view of system without thermal cover. Pressure vessel is below vacuum path and in oil bath. External electronic racks not shown.

- 1) Laser.
- 2) Beam splitter (50% transmission).
- 3) Coupling plate with mirror "B" (59.% transmission).
- 4) Coupling plate with mirror "A" (0.04% transmission).
- 5) Vacuum path between mirrors "A" and "B".

- 6) Location of fringe image and photodetectors.
- 7) Fringe photodetector preamplifier.
- 8) Translation stages.
- 9) LVDT for intermediate reference length.

Figure 6: Schematic drawing of pressure (in excess of atmospheric pressure) versus fractional change of manganin resistance, where ΔP_M is the maximum deviation of the manganin gauge from linearity between P_1 and P_{Hg} .

Figure 1

"H" shape core.

Core length change correction known and small. Requires spring loading.

Indicates magnetically soft ferromagnetic material.

Figure 2

Figure 3

FRINGE CIRCLE PRODUCED BY A MODULATION.
(TOTAL λ SWEEP $\sim 3. \times 10^{-7} \lambda_0$ AT 60Hz.)

FRINGE CIRCLE PRODUCED BY RELATIVE
MOTION OF MIRRORS "A" AND "B".
(~ 35 . FRINGES PER SECOND)

FRINGE TRACE WITH OIL CIRCULATION
SYSTEM ON.

FRINGE TRACE WITH OIL CIRCULATION
SYSTEM OFF.

Figure 4

Figure 5

Figure 6