

# Temperature Dependence of the Velocity Derivatives of Periclase<sup>1</sup>

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The temperature dependence of the sound velocity in polycrystalline MgO has been determined from +80 to -80°C at atmospheric pressure and to 2 kb at -78.5°C. These results are compared with previous measurements to 4 kb at 34.6°C. From these measurements the critical temperature gradient  $(\partial T/\partial P)_c$  for MgO was determined. Because  $(\partial T/\partial P)_c$  was found to be greater for *P* waves than for *S* waves in MgO, and both are small compared with previously reported values of  $(\partial T/\partial P)_c$  for the earth, it is possible to explain the marked presence of a low-velocity zone for *S* waves and the absence of a low-velocity zone for *P* waves in a homogeneous earth.

## INTRODUCTION

In a previous paper [Anderson and Schreiber, 1965] the pressure derivatives of the elastic constants of polycrystalline MgO, measured at a pressure of 4 kb at 34.6°C, were reported. Many applications of those data to geophysical problems were indicated, including the method of estimating the volume up to pressures of 100 kb. These data were obtained and reported because recent publications of petrologic models of the deep earth, in which MgO is required deep in the mantle, suggested the need for basic experimental data that could be used in evaluating these models [Sclar *et al.*, 1964; McQueen *et al.*, 1964].

In this communication we continue our report of the data on MgO and in particular (1) report the dependence of the sound velocities and bulk modulus upon temperature between -78.5°C and +80°C; (2) report the value of the pressure derivative of the bulk modulus  $dB/dP$  at -78.5°C and the value of the derivative of temperature with depth at constant velocity and compare these with those obtained at room temperatures; and (3) comment upon how these values relate to a few of the outstanding problems connected with the earth's interior.

## EXPERIMENTAL PROCEDURE

**MgO specimen.** The polycrystalline MgO specimen was the same as the one that was

used in our previous experiments. It was prepared by the hot-pressing technique and produced at the University of California at Berkeley. This specimen is of exceptional quality. It is transparent in the visible region, yields the same infrared reflection spectrum as the single crystal, and is 99.9+% of theoretical density. The size of the specimen is about 1.2 cm on edge and 0.6 cm in the direction of sound propagation. The sample was checked by rotating the driver crystal on the specimen surface and found to be isotropic for shear waves.

**Temperature variations.** Temperature was measured with a copper-constantan thermocouple, placed in contact with the specimen within the pressure vessel. The thermocouple output was measured with a Leeds & Northrup K-3 potentiometer and an electronic null detector. Temperatures above ambient were obtained by thermostating the vessel. For low-temperature runs, the vessel was cooled by placing it in a polystyrene insulating container and gradually adding dry ice. The rate of cooling was about 1°C every 20 minutes. This low rate was necessary to ensure that a steady state had been reached for each velocity measurement. Temperature variation was less than 0.1°C during the velocity determination at each temperature point.

**Pressure measurement.** The pressure system was the same as in our previous study (1965). Pressure was monitored with a Heise gage and measured with a Harwood 0- to 60,000-lb/in.<sup>2</sup> pressure balance. Velocities were determined at 2000-lb/in.<sup>2</sup> pressure increments, and only when

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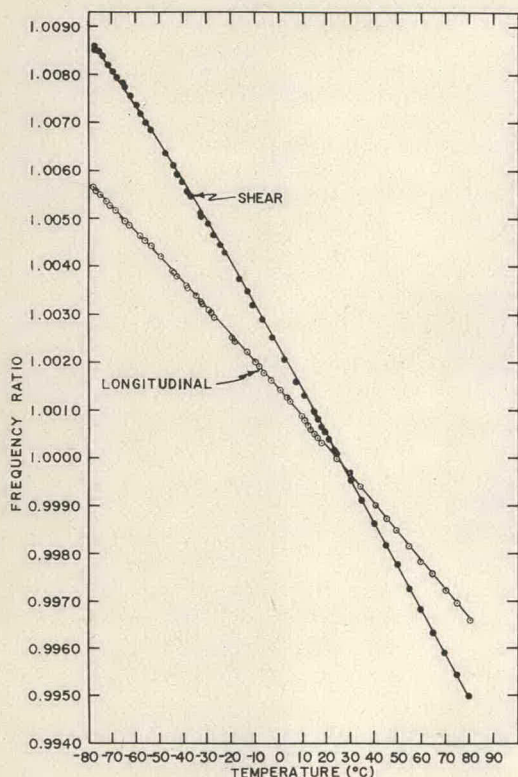


Fig. 1. Variation of the frequency ratio  $F/F_0$  with temperature from +80 to -80°C.

the pressure balance was at equilibrium pressure. During the pressure runs the temperature of the specimen was thermostated at the 1-atm sublimation temperature of dry ice (-78.5°C). Compression of the pressure-transmitting fluid (nitrogen) raised the temperature, and waiting periods of 15 to 20 minutes were required. During the pressure runs, all velocity measurements were made at a temperature held within  $\pm 0.05^\circ\text{C}$  of the reference temperature.

**Velocity determinations.** The velocity as a function of temperature and pressure was measured by ultrasonic interferometry using the technique of pulse superposition. This method has been described and its accuracy discussed in detail by its originator [McSkimin, 1961, 1964; McSkimin and Andreatch, 1962, 1963]. This technique readily permits measurement of time to better than 1 part in 100,000 and changes in time to intervals of 0.1 nsec. The details of the method will not be discussed here, but the equations will be outlined. The time delay within the specimen,  $\tau$ , is very nearly the period of the measured pulse repetition frequency  $F$  and is given by

$$p\tau = 1/F + p\theta/2\pi f - n/f \quad (1)$$

where  $p$  and  $n$  are experimentally determinable constants,  $\theta$  is a correction for the time delay

TABLE 1. Velocity and Bulk Modulus with Temperatures at 1 Atmosphere

| Temperature, °C | $v_p$ , km/sec | $v_s$ , km/sec | $\rho$ , g/cm <sup>3</sup> | $B_s$ , kb | $\alpha^* \times 10^6$ , °C <sup>-1</sup> |
|-----------------|----------------|----------------|----------------------------|------------|-------------------------------------------|
| -80             | 9.8186         | 6.0145         | 3.5904                     | 1729.6     | 7.30                                      |
| -78.5           | 9.8179         | 6.0139         | 3.5903                     | 1729.4     | 7.36                                      |
| -70             | 9.8143         | 6.0109         | 3.5896                     | 1728.3     | 7.72                                      |
| -60             | 9.8100         | 6.0069         | 3.5888                     | 1727.2     | 8.08                                      |
| -50             | 9.8057         | 6.0027         | 3.5879                     | 1726.1     | 8.43                                      |
| -40             | 9.8014         | 5.9982         | 3.5870                     | 1725.2     | 8.76                                      |
| -30             | 9.7969         | 5.9938         | 3.5861                     | 1724.1     | 9.09                                      |
| -20             | 9.7924         | 5.9893         | 3.5851                     | 1723.1     | 9.40                                      |
| -10             | 9.7877         | 5.9847         | 3.5841                     | 1721.9     | 9.68                                      |
| 0               | 9.7830         | 5.9801         | 3.5830                     | 1720.7     | 9.94                                      |
| 10              | 9.7783         | 5.9754         | 3.5819                     | 1719.6     | 10.18                                     |
| 20              | 9.7736         | 5.9706         | 3.5808                     | 1718.5     | 10.40                                     |
| 25              | 9.7711         | 5.9682         | 3.5803                     | 1717.9     | 10.51                                     |
| 30              | 9.7686         | 5.9658         | 3.5797                     | 1717.2     | 10.61                                     |
| 40              | 9.7636         | 5.9610         | 3.5786                     | 1715.9     | 10.77                                     |
| 50              | 9.7586         | 5.9562         | 3.5774                     | 1714.5     | 10.96                                     |
| 60              | 9.7536         | 5.9515         | 3.5762                     | 1713.2     | 11.12                                     |
| 70              | 9.7487         | 5.9467         | 3.5750                     | 1711.9     | 11.24                                     |
| 80              | 9.7437         | 5.9420         | 3.5738                     | 1710.6     | 11.36                                     |

\*  $\alpha$  is smoothed data as read from our plot of the data of Ganesan [1962], Sharma [1950], and Skinner [1957].

introduced by the transducer-specimen bond, and  $f$  is the carrier frequency within the pulse ( $2$  to  $6 \times 10^7$  cps). Operation is such that  $p$  and  $n$  are generally chosen to be  $1$  and  $0$ , respectively, thus simplifying (1).

The velocity  $v$ , as a function of either pressure or temperature, is determined from

$$v = v_0(F/F_0)(l/l_0) \quad (2)$$

where the subscripts refer to the initial conditions ( $1$  atm, room temperature) at which the velocity  $v_0$  has been carefully determined. The ratio  $F/F_0$  is obtained from the experiment. The length ratio  $l/l_0$  is calculated from thermal expansion data for the temperature runs at constant pressure; for the pressure runs at constant temperature,  $l/l_0$  is computed from the frequency data as described by Cook [1957].

#### EXPERIMENTAL RESULTS

*Variation with temperature at 1 atmosphere.* The variation of the frequency ratio  $F/F_0$  with temperature from  $+80$  to  $-80^\circ\text{C}$  is shown in Figure 1 for both shear and longitudinal modes. The ratio  $l/l_0$  was determined from the thermal expansion data of Ganesan [1962], Skinner [1957], and Sharma [1950]. The velocity was computed at a number of temperatures from (2), and the density calculated from

$$\rho = \rho_0(l_0/l)^3 \quad (3)$$

where  $\rho_0 = 3.5803$  g/cm<sup>3</sup> is the measured density at  $25^\circ\text{C}$ . From these data the bulk modulus was computed at the same temperatures (see Table 1). Figure 2 is a plot of the first derivative of the bulk modulus with temperature. We find this to vary quite slowly with temperature,  $(\partial B_s/\partial T)_P$  going from  $0.13$  kb/ $^\circ\text{C}$  at room temperature to  $0.11$  kb at the temperature of

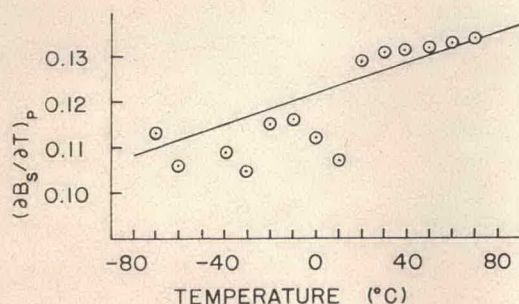


Fig. 2. Plot of the first derivative of the bulk modulus with temperature.

TABLE 2. Frequency Ratio and Length Ratio versus Pressure at  $-78.5^\circ\text{C}$

| Pressure, kb | $F/F_0$   |              |            |
|--------------|-----------|--------------|------------|
|              | Shear     | Longitudinal | $l_0/l^*$  |
| 0            | 1.000 000 | 1.000 000    | 1.000 0000 |
| 0.5          | 1.000 451 | 1.000 496    | 1.000 1096 |
| 1.0          | 1.000 903 | 1.000 992    | 1.000 2192 |
| 1.5          | 1.001 355 | 1.001 488    | 1.000 3288 |
| 2.0          | 1.001 806 | 1.001 984    | 1.000 4383 |

\* Calculated according to Cook [1957].

dry ice. To compute the variation of bulk modulus with temperature, we differentiate

$$B_s = \rho[v_p^2 - (4/3)v_s^2] \quad (4)$$

with respect to temperature and obtain

$$\left(\frac{\partial B_s}{\partial T}\right)_P = 2\rho\left[v_p\left(\frac{\partial v_p}{\partial T}\right)_P - \frac{4}{3}v_s\left(\frac{\partial v_s}{\partial T}\right)_P\right] - \alpha_v B_s \quad (5)$$

which was used in computing the values plotted in Figure 2. In (5),  $\alpha_v$  is the volume expansion.

*Variation with pressure.* From the data in Tables 2 and 3, at  $-78.5^\circ\text{C}$  the variation of velocity with pressure was found to be

$$v_p = 9.8179 + 7.57 \times 10^{-3}P \quad (6)$$

$$v_s = 6.0138 + 4.07 \times 10^{-3}P$$

where velocities are in kilometers per second and pressure is in kilobars.

The variation of the adiabatic bulk modulus with pressure was computed from the velocity data using

$$\left(\frac{\partial B_s}{\partial P}\right)_T = 2\rho\left[v_p\left(\frac{\partial v_p}{\partial P}\right)_T - \frac{4}{3}v_s\left(\frac{\partial v_s}{\partial P}\right)_T\right] + 1 + \alpha_v \gamma T \quad (7)$$

TABLE 3. Velocities and Bulk Modulus versus Pressure for Polycrystalline MgO at  $-78.5^\circ\text{C}$

| Pressure | Density, g/cm <sup>3</sup> | $v_p$ , km/sec | $v_s$ , km/sec | $B_s$ , kb |
|----------|----------------------------|----------------|----------------|------------|
| 0.0      | 3.5903                     | 9.8179         | 6.0139         | 1729.4     |
| 0.5      | 3.5914                     | 9.8216         | 6.0158         | 1731.4     |
| 1.0      | 3.5926                     | 9.8254         | 6.0179         | 1733.5     |
| 1.5      | 3.5938                     | 9.8292         | 6.0199         | 1735.6     |
| 2.0      | 3.5949                     | 9.8330         | 6.0220         | 1737.6     |

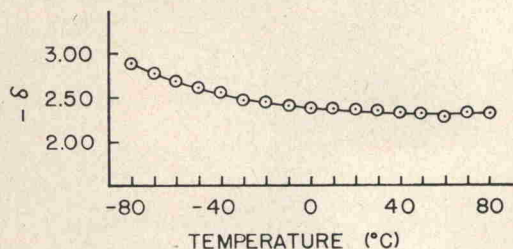


Fig. 3. Plot of  $\delta$  as a function of temperature.

where  $\gamma$  is the Grüneisen constant ( $\gamma = 1.67$ ), evaluated later. Equation 7 is obtained by differentiating (4) with pressure. This leads to a value of  $(\partial B_s/\partial P)_T = 4.00$  at  $-78.5^\circ\text{C}$ , which is comparable to a value of 3.92 at  $34.6^\circ\text{C}$ .

Another function which is due to Grüneisen [1926] can be computed also. This function, which we call  $\delta$ , is defined as

$$\delta = -\frac{1}{B_s} \left( \frac{\partial B_s}{\partial T} \right)_P \bigg/ \frac{1}{V} \left( \frac{\partial V}{\partial T} \right)_P \quad (8)$$

and should be constant with temperature at higher temperatures. This function, then, may provide a useful means of estimating the value of the temperature derivative of the adiabatic bulk modulus at the elevated temperatures within the earth's interior from experiments over a narrow temperature range through

$$(\partial B_s/\partial T)_P = -\delta \gamma C_{pP} \quad (9)$$

Even at these low temperatures, the tendency of  $\delta$  to approach an asymptotic value at elevated temperature can be seen (Figure 3).

**Isothermal values.** The values of interest to geophysicists are the isothermal moduli, and it is necessary to compute these from the adiabatic values obtained from our experiment. This computation involves a knowledge of  $\gamma$ ,  $\alpha_v = 3\alpha$ , and the absolute temperature. The value of  $\gamma$  can be calculated at  $-78.5^\circ\text{C}$ . We have  $\alpha_v = 22.08 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$  [Ganesan, 1962]; the specific heat capacity  $C_p = 6.37 \times 10^6 \text{ ergs/g } ^\circ\text{C}$  [Baron, 1959];  $T = 194^\circ\text{K}$ ,  $B_s = 1729.4$ , and  $\rho = 3.5903 \text{ g/cm}^3$ . From

$$\gamma = \alpha_v B_s / \rho C_p \quad (10)$$

we obtain  $\gamma = 1.67$  for MgO at  $-78.5^\circ\text{C}$ ,  $P = 1 \text{ atm}$ .

The value of the isothermal bulk modulus is calculated from

$$B_T = B_s(1 + T\alpha_v\gamma)^{-1} \quad (11)$$

whence we obtain  $B_T = 1717.1 \text{ kb}$  at  $-78.5^\circ\text{C}$  and 1 atm.

The isothermal pressure derivative of the bulk modulus is obtained from

$$\begin{aligned} \left( \frac{\partial B_T}{\partial P} \right)_T &= \left( \frac{\partial B_s}{\partial P} \right)_T \\ &+ T\alpha_v\gamma \left( \frac{C_p}{C_p} \right) \left[ \frac{-2}{\alpha_v B_T} \left( \frac{\partial B_T}{\partial T} \right)_P - 2 \left( \frac{\partial B_s}{\partial T} \right)_P \right] \\ &+ \left[ T\alpha_v\gamma \left( \frac{C_p}{C_p} \right) \right]^2 \left[ \left( \frac{\partial B_s}{\partial P} \right)_T - \frac{1}{\alpha_v^2} \left( \frac{\partial \alpha_v}{\partial T} \right) - 1 \right] \end{aligned} \quad (12)$$

The values of the correction terms used in (12) are listed in Table 4.

To compute the change of isothermal modulus with temperature, we differentiate (11) with respect to temperature and obtain

$$\begin{aligned} \left( \frac{\partial B_T}{\partial T} \right)_P &= \left( \frac{\partial B_s}{\partial T} \right)_P (1 + T\alpha_v\gamma)^{-1} \\ &- \frac{B_s\gamma}{(1 + T\alpha_v\gamma)^2} \left[ \alpha_v + T \left( \frac{\partial \alpha_v}{\partial T} \right)_P \right] \end{aligned} \quad (13)$$

The results of these experiments at  $-78.5^\circ\text{C}$  are compared with the results previously reported for  $34.6^\circ\text{C}$  in Table 5.

## DISCUSSION

The data presented here, in conjunction with our earlier data [Anderson and Schreiber, 1965] can be used in estimating the critical temperature gradient of MgO. The critical temperature gradient [Birch, 1958] is that value of  $(\partial T/\partial P)_v$  which when applied to the material causes the sound velocity to be refracted with positive curvature. Thus, if the critical temperature gradient is exceeded, the appropriate sound velocity will decrease with increasing pressure (or depth, in the case of the earth). We comment in passing that MgO is probably not in the upper mantle, and so our results do not apply directly to the low-velocity zone. We wish to emphasize, however, that similar results are likely to be found for the actual constituents of the upper mantle. The critical gradients for the material are given by

$$(\partial T/\partial P)_v = -\frac{(\partial v/\partial P)_T}{(\partial v/\partial T)_P} \quad (14)$$

TABLE 4. Corrections for Computing  $\partial B_T/\partial P$  at  $-78.5^\circ\text{C}$ 

| Condition  | Symbol                                          | Amount                   | Units                    |
|------------|-------------------------------------------------|--------------------------|--------------------------|
| Measured   | $(\partial B_s/\partial P)_T$                   | 4.00                     |                          |
| Measured   | $\alpha_s^*$                                    | $22.08 \times 10^{-6}$   | $^\circ\text{C}^{-1}$    |
| Measured   | $C_P^*$                                         | $6.37 \times 10^6$       | ergs/g/ $^\circ\text{C}$ |
| Measured   | $\chi_s^*\dagger$                               | $5.7823 \times 10^{-13}$ | $\text{cm}^2/\text{g}$   |
| Calculated | $\gamma^*$                                      | 1.67                     |                          |
| Calculated | $\chi_T\dagger$                                 | $5.8238 \times 10^{-13}$ | $\text{cm}^2/\text{g}$   |
| Measured   | $(\partial B_T/\partial T)_P$                   | -0.25                    | kb/ $^\circ\text{K}$     |
| Measured   | $(\partial \alpha_s/\partial T)_P$              | $1.3 \times 10^{-7}$     |                          |
| Calculated | $1 + \alpha_s \gamma T$                         | 1.00717                  |                          |
| Calculated | $\alpha_s \gamma T (C_V/C_P)$                   | $7.11 \times 10^{-3}$    |                          |
| Calculated | $2(\alpha B_T)^{-1}(\partial B_T/\partial T)_P$ | -12.9                    |                          |
| Calculated | $\alpha_s^{-2}(\partial \alpha_s/\partial T)_P$ | 266                      |                          |
| Calculated | Total 2nd term, eq. 12                          | 0.034                    |                          |
| Calculated | Total 3rd term, eq. 12                          | -0.013                   |                          |
| Calculated | $(\partial B_T/\partial P)_T$                   | 4.02                     |                          |

\* At  $-78.5^\circ\text{C}$ . $\dagger \chi_s$  and  $\chi_T$  are the adiabatic and isothermal compressibility.

where  $v$  is the shear or longitudinal velocity.

From Table 5 we obtain the values of  $(\partial v/\partial P)_T$  and  $(\partial v/\partial T)_P$  and from these we obtain values of  $(\partial T/\partial P)_v$  at the two temperatures:

|                                 | $-78.5^\circ\text{C}$ | $34.6^\circ\text{C}$ |                            |
|---------------------------------|-----------------------|----------------------|----------------------------|
| $(\partial T/\partial P)_{v_s}$ | 17.6                  | 15.4                 | $^\circ\text{C}/\text{kb}$ |
| $\partial T/\partial P)_{v_s}$  | 11.3                  | 9.08                 | $^\circ\text{C}/\text{kb}$ |

Thus  $(\partial^2 T/\partial P \partial T)$  is about  $-0.02$  deg/kb at low temperature and is probably smaller at high

temperature; therefore the second derivative does not alter the main argument. In trying to assess, for example, whether a material such as MgO is a dominant component of the earth's mantle, we compare the above values with the geothermal gradient, given by  $(\partial T/\partial P)_{\text{earth}}$ .

Whenever  $(\partial T/\partial P)_{\text{earth}} > (\partial T/\partial P)_{v_s}$ , it follows that velocity decreases with depth, i.e., that  $dv/dZ$  is negative. The lower the value of the critical temperature gradient of materials which constitute the earth, the easier it is to explain a

TABLE 5. Comparison of Parameters Determined at  $-78.5^\circ\text{C}$  and  $34.6^\circ\text{C}$ 

| Parameter                         | Temperature           |                        | Units                      |
|-----------------------------------|-----------------------|------------------------|----------------------------|
|                                   | $-78.5^\circ\text{C}$ | $34.6^\circ\text{C}$   |                            |
| $v_P$ at $P = 0$                  | 9.8179                | 9.7662                 | km/sec                     |
| $v_s$ at $P = 0$                  | 6.0138                | 5.9635                 | km/sec                     |
| $(\partial v_P/\partial P)_T$     | $7.57 \times 10^{-3}$ | $7.711 \times 10^{-3}$ | km/sec/kb                  |
| $(\partial v_s/\partial P)_T$     | $4.07 \times 10^{-3}$ | $4.351 \times 10^{-3}$ | km/sec/kb                  |
| $(\partial v_P/\partial T)_{P=0}$ | $4.3 \times 10^{-4}$  | $5.0 \times 10^{-4}$   | km/sec/ $^\circ\text{C}$   |
| $(\partial v_s/\partial T)_{P=0}$ | $3.6 \times 10^{-4}$  | $4.8 \times 10^{-4}$   | km/sec/ $^\circ\text{C}$   |
| $B_s, P = 0$                      | 1729.4                | 1717.0                 | kb                         |
| $B_T, P = 0$                      | 1717.1                | 1691.0                 | kb                         |
| $(\partial B_s/\partial P)_T$     | 4.00                  | 3.92                   |                            |
| $(\partial B_T/\partial P)_T$     | 4.02                  | 3.94                   |                            |
| $(\partial B_s/\partial T)_{P=0}$ | -0.11                 | -0.13                  | kb/ $^\circ\text{C}$       |
| $(\partial B_T/\partial T)_{P=0}$ | -0.25                 | -0.29                  | kb/ $^\circ\text{C}$       |
| $\gamma$                          | 1.67                  | 1.60                   |                            |
| Poisson's ratio                   | 0.200                 | 0.203                  |                            |
| $(\partial T/\partial P)_{v_P}$   | 17.6                  | 15.4                   | $^\circ\text{C}/\text{kb}$ |
| $(\partial T/\partial P)_{v_s}$   | 11.3                  | 9.08                   | $^\circ\text{C}/\text{kb}$ |
| Density                           | 3.5903                | 3.5800                 | $\text{g}/\text{cm}^3$     |

low-velocity zone in the upper mantle. Furthermore, if  $(\partial T/\partial P)_{\text{P}}$  is greater than  $(\partial T/\partial P)_{\text{S}}$ , it is possible for the geothermal gradient to produce a  $dv_{\text{P}}/dZ$  that is negative while producing a  $dv_{\text{S}}/dZ$  that is zero or positive. We therefore attach significance to the fact that, for MgO, the critical temperature gradient is 70% more for the  $P$  wave than for the  $S$  wave; further, both values of the critical gradient are low, 4.4°C/km for  $P$  waves and 2.7°C/km for  $S$  waves (using the 34.6°C data and 3.3 km/kb for the earth's pressure gradient).

It may turn out that when accurate measurements are made of  $(\partial T/\partial P)_{\text{P}}$  for other materials related to the earth's mantle, the value will also be low, as we have found for MgO. Such a result would have considerable effect on our thinking about the composition of the mantle on the basis of petrologic models [Ringwood, 1962; Clark and Ringwood, 1964] because the investigators have relied, in general, on estimates of the critical gradient which are larger than those reported here. There are data on  $\text{Al}_2\text{O}_3$  [Schreiber and Anderson, 1966] for which  $(\partial P/\partial T)_{\text{P}} = 3.5^\circ\text{C}/\text{km}$  and  $(\partial P/\partial T)_{\text{S}} = 2.1^\circ\text{C}/\text{km}$ , which is less than for MgO. Thus the evidence on oxides to date implies that the geothermal gradient is quite low.

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