# THERMODYNAMIC PROPERTIES OF THE GENERALIZED MURNAGHAN EQUATION OF STATE OF SOLIDS

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**Abstract.** A relation between the pressure (P) and the volume (V) of solids was obtained by Murnaghan by assuming that the isothermal bulk modulus (B) varies linearly with P. The equation was derived for isothermal conditions but in order to treat, e.g. the thermal expansivity ($\alpha$) at high P, it is necessary to let its parameters vary with temperature (T). We study the thermodynamic properties of the most general form of the Murnaghan equation, where all its parameters- viz. V and B at P = 0, and $(\partial B/\partial P)_T$- are functions of T. We obtain general relations for the effect of P upon several thermodynamic quantities, viz. $\alpha$, the product $\alpha B$, the Anderson-Grüneisen parameter ($\delta$) and the Grüneisen parameter ($\gamma_G$). We show that certain combinations of those parameters are given by the Murnaghan approximation a simple dependence on P. Moreover, we report a relation between $\alpha$, $\delta$ and V that generalizes a previous result by us [J.Phys.Chem.Solids 47:605 (1986)], and the Anderson equation for expansivity at high P.

Keywords: Anderson-Grüneisen, bulk modulus, equation of state, expansivity, high pressure, Murnaghan.

## 1. INTRODUCTION

In the Murnaghan [1] approximation to the isothermal equation of state of solids a relation between pressure (P) and volume (V) is derived from the assumption that the isothermal bulk modulus (B) varies linearly with P. The Murnaghan equation has been used in the representation of isothermal P-V data for solids, and in the extrapolation of low-pressure information to large compressions [2]. The possibility of generalizing the equation by including some temperature effects was considered long ago by Gilvarry [3]. More recently, a temperature-dependent form of the Murnaghan equation of state was proposed [4], which was based on accounting for the variation with temperature (T) of the volume and bulk modulus at zero pressure. The equation was used in treating the effect of pressure upon the Gibbs energy of the condensed phases of various metals [5,6], the thermal expansivity and the Anderson-Grüneisen parameter [7] at high pressures, and in a thermodynamic analysis of shock-wave data [8].

In the present work we shall go on a step further and let the remaining parameter in the Murnaghan equation, viz. the pressure-derivative of the isothermal bulk modulus, vary with T. The possibility of that variation was mentioned by Birch [9] in a discussion of the thermal expansivity at high pressures, but a detailed analysis of its effects on the thermodynamic parameters of the solid seems to be lacking. Such an analysis is reported in the present paper, which is organized as follows. In Section 2 we derive the basic relations of the paper, concerning the behavior of the isothermal bulk modulus and the thermal expansion coefficient. In Section 3 we use those relations to study the

behavior of various thermodynamic quantities, viz. the product $\alpha B$, which is related to the thermal pressure of solids, the Anderson-Grüneisen parameter, which is involved in relations between expansivity and volume at high pressures, and the thermodynamic Grüneisen parameter. In Section 4 we summarize the work and give our conclusions.

## 2. BASIC THERMODYNAMIC RELATIONS

### 2.1 Isothermal Bulk Modulus

We express the isothermal bulk modulus B

$$B(T,P) = -V\left(\frac{\partial P}{\partial V}\right)_T \tag{1}$$

as a linear function of P

$$B(T,P) = B(T,0) + n\,P \tag{2}$$

where $B(T,0)$ is the bulk modulus at zero pressure and the pressure-independent parameter n is a function of temperature. Derivating Eq.(2) with respect to T yields

$$\left(\frac{\partial B}{\partial T}\right)_P = \frac{dB(T,0)}{dT} + \left(\frac{dn}{dT}\right)P \tag{3}$$

which can be written as

$$\left(\frac{\partial B}{\partial T}\right)_P = -\,\alpha(T,0)B(T,0)\left[\delta(T,0)-\beta(T,0)\,\frac{P}{B(T,0)}\right] \tag{4}$$

where $\alpha$ represents the thermal expansion coefficient,

$$\alpha(T,P) = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P \qquad (5)$$

$\delta$ is the so-called Anderson-Grüneisen parameter [10],

$$\delta(T,P) = -\frac{1}{\alpha B}\left(\frac{\partial B}{\partial T}\right)_P \qquad (6)$$

and $\beta(T,0)$ is a dimensionless quantity introduced by us,

$$\beta(T,0) \equiv \frac{1}{\alpha(T,0)}\left(\frac{dn}{dT}\right) \qquad (7)$$

According to Eq.(4) the effect of temperature upon B(T,P) depends on the parameters $\delta(T,0)$ and $\beta(T,0)$. In the most frequent case B(T,0) decreases with increasing temperature and $\alpha(T,0)$ is positive, i.e. $\delta(T,0)$ is a positive quantity. Some insight on the behavior of $\beta(T,0)$ may be gained from information on the temperature dependence of $(\partial B/\partial P)_T$, which is available for solids of various types. For instance, data on solid Ne, Ar, Kr and Xe, analysed by Birch [11] show an increase with T of the pressure derivative of B at P = 0, and a similar behavior is suggested by ultrasonic measurements on NaCl [12] between 300 and 800K, and on $CaF_2$ [13] above 195K. Ultrasonic measurements on Cu [14] at 77 and 295K suggest a decrease of n with increasing temperature. Contrasting with that, a thermodynamic study [15] of shock-wave information for Mo, comprising Hugoniot data obtained starting at 293K [16] and 1673K [17], based on the method of analysis presented in Ref.8 and leting n vary linearly with T above room-temperature, indicates that $dn/dT > 0$. In view of these results we shall not make any assumption about the sign of dn/dT and $\beta(T,0)$, but treat them as quantities that, in principle, can

have either sign.

For the particular case where $\beta(T,0) > 0$ and -as usual- $\alpha(T,0)$ and $\delta(T,0)$ are positive, Eq. (4) predicts that the rate of decrease of B with increasing T gets smaller at higher pressures, and becomes zero at the pressure

$$P^* = \frac{\delta(T,0)}{\beta(T,0)} B(T,0) \qquad (8)$$

The consequences of this result on the behavior of the thermal expansivity at high P are examined in the next section.

## 2.2 Thermal Expansion Coefficient

Integrating Eq. (2) at constant temperature yields

$$\frac{V(T,P)}{V(T,0)} = \left[1 + n \frac{P}{B(T,0)}\right]^{-1/n} \qquad (9)$$

which leads to the following relation for the thermal expansion coefficient $\alpha$ (Eq. (5))

$$\frac{\alpha(T,P)}{\alpha(T,0)} = \frac{1+\left[n-\delta(T,0)\right]\frac{P}{B(T,0)}}{1+ n \frac{P}{B(T,0)}} + \beta(T,0)\ \varphi_1(T,P) \qquad (10)$$

where $\delta$ and $\beta(T,0)$ are defined by Eqs. (6) and (7), respectively, and

$$\varphi_1(T,P) \equiv \frac{1}{n^2}\left\{\ln\left[1+ n \frac{P}{B(T,0)}\right] - \frac{n \frac{P}{B(T,0)}}{1+ n \frac{P}{B(T,0)}}\right\} \qquad (11)$$

Equation 10 describes the effect of pressure on $\alpha(T,P)/\alpha(T,0)$

as the sum of two terms. The first term does not involve $\beta(T,0)$, but $\delta(T,0)$. In the usual case is $\delta(T,0) > 0$, and that term decreases with increasing pressure from the value 1 at $P = 0$, down to the value $[n-\delta(T,0)]/\, n = 1 - [\delta(T,0)/\, n]$ at extremely high P. The second term in Eq.(10) is the product of the purely temperature-dependent quantity $\beta(T,0)$ times $\varphi_1$, which is the function of T and P defined by Eq.(11). In Fig.1 we plot $\varphi_1(T,P)$ versus $P/B(T,0)$, for various values of n. Since $\varphi_1(T,P)$ is positive and increases with $P/B(T,0)$, the second term in Eq.(10) will give a positive contribution to $\alpha(T,P)/\, \alpha(T,0)$ if $\beta(T,0) > 0$, and a negative contribution if $\beta(T,0) < 0$. As a consequence, in the case where $\beta(T,0) > 0$ and $\alpha(T,0) > 0$ the variation of $\alpha(T,P)$ with P will be determined by the competing effects of the two terms in Eq.(10), and there is the possibility of a minimum in the $\alpha(T,P)$ versus P function. We examine this possibility further by considering the pressure derivative of $\alpha$. The Maxwell relation

$$\left[\frac{\partial \alpha}{\partial P}\right]_T = \frac{1}{B^2}\left[\frac{\partial B}{\partial T}\right]_P \qquad (12)$$

implies that the minimum in $\alpha$ occurs at a pressure such that $(\partial B/\partial T)_P$ is zero, which corresponds to the critical pressure $P^*$ introduced in the previous section (Eq.(8)). By inserting this pressure value in Eq.(10) we evaluate the minimum $\alpha(T,P)/\alpha(T,0)$ ratio as

$$\frac{\alpha(T,P^*)}{\alpha(T,0)} = \frac{n - \delta(T,0)}{n} + \frac{\beta(T,0)}{n^2} \ln\left[1 + n\,\frac{\delta(T,0)}{\beta(T,0)}\right] \qquad (13)$$

The first term in Eq.(13) is the limiting $\alpha(T,P)/\alpha(T,0)$ ratio

given by the Murnaghan approximation for $P \rightarrow \infty$ when n is independent of T [7]. That ratio is negative if $n < \delta(T,0)$. In spite of that, Eq.(13) predicts that

$$\frac{\alpha(T,P^*)}{\alpha(T,0)} \geq 0 \qquad (14)$$

if the ratio $n/\delta(T,0)$, which is less than unity, is large enough to satisfy the relation

$$\frac{n}{\delta(T,0)} \geq 1 - \frac{\ln\left[1+ n \frac{\delta(T,0)}{\beta(T,0)}\right]}{n \frac{\delta(T,0)}{\beta(T,0)}} \qquad (15)$$

Values of the $n/\delta(T,0)$ ratio satisfying Eq.(15) with the equality sign are plotted in Fig.2 versus $\delta(T,0)/\beta(T,0)$ for various values of n. Taking, for instance, $\delta(T,P)/\beta(T,0) = 1$ and $n = 4$, we have from Fig.2 that $\alpha(T,P^*)/\alpha(T,0) = 0$ if $n/\delta(T,0) \simeq 0.6$. Larger values of $n/\delta(T,0)$ and the same $\delta(T,0)/\beta(T,0)$ will make $\alpha(T,P^*)/\alpha(T,0) > 0$, which in the usual case $\alpha(T,0) > 0$ implies that $\alpha(T,P)$ will be positive at all pressures. According to Fig.2, the smallest $n/\delta(T,0)$ ratios leading to $\alpha(T,P^*) > 0$ for other values of n decrease rapidly with the relation $\delta(T,0)/\beta(T,0)$, in particular, if $\delta(T,0)/\beta(T,0) < 3$. In Sect.3.5 of this paper we show that the condition $\delta(T,0)/\beta(T,0) = 1/n$ leads to a particularly simple expression for the volume dependence of the thermodynamic Grüneisen parameter in the Murnaghan approximation. Using Eq.(15) we predict for that case $\alpha(T,P^*)/\alpha(T,0) > 0$ if $n/\delta(T,0) > 1-\ln 2 \simeq 0.31$.

## 3. APPLICATIONS TO THERMODYNAMIC PARAMETERS OF SOLIDS

### 3.1. Pressure Effects on the Quantity αB.

The properties of the product αB are of interest in analysing the temperature dependence of the so-called thermal pressure of solids. That dependence is usually described by means of the quantity $(\partial P/\partial T)_v$, which is related to the product αB by the identity

$$\left(\frac{\partial P}{\partial T}\right)_v = \alpha B \tag{16}$$

Here we derive a relation describing the effect of P upon αB. By combining Eqs. (2) and (10) we obtain

$$\frac{\alpha(T,P)B(T,P)}{\alpha(T,0)B(T,0)} = 1+\left[n-\delta(T,0)\right]\frac{P}{B(T,0)} + \beta(T,0)\ \varphi_2(T,P) \tag{17}$$

where

$$\varphi_2(T,P) \equiv \left[1+ n\ \frac{P}{B(T,0)}\right]\ \varphi_1(T,P) \tag{18}$$

and $\varphi_1(T,P)$ is defined by Eq. (11). In Fig. 3 we plot $\varphi_2$ versus $P/B(T,0)$, for various values of n. Equation (17) describes the pressure dependence of αB as the sum of two contributions. The first contribution is proportional to P, and is positive if $n > \delta(T,0)$ and negative of $n < \delta(T,0)$. The second contribution is the product of the quantity $\varphi_2$, which is positive and increases with P (Fig. (3)), times the parameter $\beta(T,0)$, which measures the temperature dependence of n (Eq. (7)) and can have either sign (Sect. 2.1).

The pressure dependence of αB, as given by Eq. (17), is further discussed in Sect. 3.3.

### 3.2 Pressure Effects on the Anderson-Grüneisen Parameter.

The Anderson-Grüneisen parameter, $\delta$, which is defined by Eq.(6), is a useful quantity in studying properties related to the anharmonic behavior of solids. In the present study we have focussed on the thermodynamic relations between $\delta$ and three such properties, viz. the thermal expansion coefficient $\alpha$, the product $\alpha B$ and the thermodynamic Grüneisen parameter. We start by deriving a relation for the pressure dependence of $\delta$. Using Eqs.(4) and (17)we obtain

$$\frac{\delta(T,P)}{\delta(T,0)} = \frac{1 - \left[\frac{\beta(T,0)}{\delta(T,0)}\right]\frac{P}{B(T,0)}}{1 + \left[n - \delta(T,0)\right]\frac{P}{B(T,0)} + \beta(T,0)\,\varphi_2(T,P)} \qquad (19)$$

In connection with the modelling of the thermal expansivity at high pressures (Sect.3.4) it is sometimes [10] assumed that $\delta$ is independent of P. From Eq.(19) we conclude that such approximation will hold exactly at all pressures if the parameters of the Murnaghan equation satisfy the conditions

$$\beta(T,0) = 0 \qquad (20)$$

and

$$n = \delta \qquad (21)$$

Equation (20) implies that the pressure-independent parameter n is also independent of T, i.e. that n is a constant. It follows from Eqs.(20) and (21) that in the Murnaghan approximation the condition that $\delta$ is independent of P at all temperatures implies

that $\delta$ is a constant, identical to n, the pressure derivative of B. If $n-\delta(T,0) > 0$ and $\beta(T,0) > 0$ the approximation $\delta(T,P) = \delta(T,0)$ with $\delta(T,0) > 0$ leads to a larger $\delta(T,P)$ than given by the Murnaghan equation, and leads to a smaller $\delta(T,P)$ if $n-\delta(T,0) < 0$ and $\beta(T,0) < 0$.

### 3.3. Relations Between $\alpha B$ and $\delta$.

A general relation between $\alpha B$ and $\delta$ can be obtained by derivating the product $\alpha B$ with respect to P and taking into account Eqs.(6) and (12). We find

$$\left[\frac{\partial(\alpha B)}{\partial P}\right]_T = \alpha(T,P)\left\{\left[\frac{\partial B}{\partial P}\right]_T - \delta(T,P)\right\} \qquad (22)$$

Extrapolations of P-V-T data for solids [18] have often been based on the approximation that the product $\alpha B$ is independent of P at constant T. Equation (22) (with $\alpha(T,P) > 0$) indicates that such condition is fulfilled exactly when

$$\left[\frac{\partial B}{\partial P}\right]_T = \delta(T,P) \qquad (23)$$

i.e. the functions of T and P $(\partial B/\partial P)_T$ and $\delta(T,P)$ are the same. In the Murnaghan approximation $(\partial B/\partial P)_T$ is independent of P, and Eq.(23) requires $\delta$ to be independent of P. That will be the case if Eqs.(20) and (21) apply. If those equations do not apply, the approximation $\alpha(T,P)B(T,P) = \alpha(T,0)B(T,0)$ overestimates $\alpha(T,P)B(T,P)$ compared with the Murnaghan equation if $n-\delta(T,0)$ and $\beta(T,0)$ are negative, and underestimates $\alpha(T,P)B(T,P)$ if $n-\delta(T,0)$ and $\beta(T,0)$ are positive. If $n-\delta(T,0)$ and $\beta(T,0)$ have different

signs there is the possibility of a partial cancellation between the last two terms on the right-hand side of Eq. (17).

By combining Eqs. (17) and (19) we obtain

$$\frac{\alpha(T,P)B(T,P)\delta(T,P)}{\alpha(T,0)B(T,0)\delta(T,0)} = 1 - \left[\frac{\beta(T,0)}{\delta(T,0)}\right]\frac{P}{B(T,0)} \qquad (24)$$

Equation (24) summarizes our results about the effects of pressure upon the quantities $\alpha$, B and $\delta$ of a solid that obeys the temperature-dependent Murnaghan equation of state. It shows that the product $\alpha B\delta$ varies linearly with pressure, decreasing with increasing P if the ratio $\beta(T,0)/\delta(T,0)$ is positive, and increasing with P if the ratio is negative. When n is independent of temperature and $\alpha(T,0) \neq 0$, we have from Eq. (7) $\beta(T,0) = 0$, and Eq. (24) takes the form

$$\alpha(T,P)B(T,P)\delta(T,P) = k(T) \qquad (25)$$

where k(T) is only a function of temperature.

### 3.4 Relations Between $\alpha$, $\delta$ and V.

The analysis of the thermal expansivity at high pressures has sometimes [9,10] been made in terms of a relation between $\alpha$ and V. A relation of that kind can be obtained from the present results by starting from Eq. (24) and eliminating the ratio $B(T,P)/B(T,0)$ by means of Eqs. (2) and (9). We obtain

$$\frac{\alpha(T,P)\delta(T,P)}{V^n(T,P)} = \frac{\alpha(T,0)\delta(T,0)}{V^n(T,0)}\left\{1 - \left[\frac{\beta(T,0)}{\delta(T,0)}\right]\frac{P}{B(T,0)}\right\} \qquad (26)$$

Equation (26) is proposed here as the most general relation between $\alpha$ and V that follows from the Murnaghan approximation. If the n parameter is independent of T and $\alpha(T,0) \neq 0$, we have from Eq.(7) $\beta(T,0) = 0$, and Eq.(26) reduces to

$$\frac{\alpha(T,P)\delta(T,P)}{V^n(T,P)} = \frac{\alpha(T,0)\delta(T,0)}{V^n(T,0)} \tag{27}$$

which has previously been reported by us [7]. If the Anderson-Grüneisen parameter is independent of pressure Eqs.(20) and (21) hold, and Eq.(26) leads to

$$\frac{\alpha(T,P)}{V^n(T,P)} = \frac{\alpha(T,0)}{V^n(T,0)} \tag{28}$$

Equation (28) reproduces Anderson's [10] relation for expansivity at high pressures.

### 3.5 Volume Dependence of the Grüneisen Parameter

The thermodynamic Grüneisen parameter $\gamma_G$ is defined as

$$\gamma_G(T,P) = \frac{\alpha\, B\, V}{C_V} \tag{29}$$

where $C_V$ is the heat capacity at constant volume. The properties of $\gamma_G$ are the considerable interest in the analysis of experimental data, and discussions of the equation of state of solids. In particular, the volume dependence of $\gamma_G$ is often considered in connection with the treatment of shock-wave data [19]. In the present section we examine the relation between $\gamma_G$ and V given by the temperature-dependent Murnaghan approximation.

By combining Eqs. (24) and (29) we obtain

$$\frac{\gamma_G(T,P)C_V(T,P)\delta(T,P)}{V(T,P)} = \frac{\gamma_G(T,0)C_V(T,0)\delta(T,0)}{V(T,0)}\left\{1-\left[\frac{\beta(T,0)}{\delta(T,0)}\right]\frac{P}{B(T,0)}\right\} \quad (30)$$

Eliminating P/B(T,0) by means of eq. (9) yields

$$\frac{\gamma_G(T,P)C_V(T,P)\delta(T,P)}{\gamma_G(T,0)C_V(T,0)\delta(T,0)} = \frac{V(T,P)}{V(T,0)}\left[1-\frac{\beta(T,0)}{n\ \delta(T,0)}\left\{\left[\frac{V(T,P)}{V(T,0)}\right]^{-n}-1\right\}\right] \quad (31)$$

which gives the volume dependence of the product $\gamma_G C_V \delta$. If the n parameter is independent of T and $\alpha(T,0) \neq 0$ we have form Eq. (7) $\beta(T,0) = 0$, and Eq. (31) becomes

$$\frac{\gamma_G(T,P)C_V(T,P)\delta(T,P)}{\gamma_G(T,0)C_V(T,0)\delta(T,0)} = \frac{V(T,P)}{V(T,0)} \quad (32)$$

i.e. the product $\gamma_G C_V \delta$ is predicted as proportional to V. When n depends on T Eq. (32) describes only the limiting behavior for $V(T,P)/V(T,0) \approx 1$, but at larger compressions the predictions of Eq. (32) will overestimate the product $\gamma_G(T,P)C_V(T,P)\delta(T,P)$ if $[\beta(T,0)/n\ \delta(T,0)] > 0$, and will underestimate the product if $[\beta(T,0)/n\ \delta(T,0)] < 0$.

In the particular case where

$$\frac{\beta(T,0)}{\delta(T,0)} = n \quad (33)$$

the volume dependence of $\gamma_G$ (Eq. (31)) takes a rather simple form, viz.

$$\frac{\gamma_G(T,P)C_V(T,P)\delta(T,P)}{\gamma_G(T,0)C_V(T,0)\delta(T,0)} = \frac{V(T,P)}{V(T,0)}\left\{2 - \left[\frac{V(T,P)}{V(T,0)}\right]^{-n}\right\} \qquad (34)$$

Finally, if the Anderson-Grüneisen parameter is independent of P, Eqs.(20) and (21) hold, and Eq.(30) takes the form

$$\left[\frac{\gamma_G(T,P)}{V(T,P)}\right] \cdot C_V(T,P) = g(T) \qquad (35)$$

where g(T) is only a function of T. At a fixed temperature, $C_V$ and the ratio $(\gamma_G/V)$ are predicted to vary with pressure in such a way that their product remains constant.

## 4. SUMMARY AND CONCLUDING REMARKS

The Murnaghan approximation is a useful tool in the treatment of pressure effects on the properties of solids. In the present work we have studied the thermodynamic properties of the most general form of the Murnaghan equation, where all its parameters are temperature-dependent. We have obtained relations describing the pressure dependence of the thermal expansivity, the product $\alpha B$ and other thermodynamic quantities. Our results show that in the Murnaghan approximation certain combinations of thermodynamic parameters, viz. $\alpha B\delta$, $\alpha\delta/V^n$ and $\gamma_G C_V \delta/V$, vary in a rather simple way with pressure, and we report relations describing that variation in terms of properties corresponding to P=0. The present work also illustrates on the use of the Anderson-Grüneisen parameter in treating pressure effects upon quantities like $\alpha$, $\alpha B$ and $\gamma_G$, that are related to the anharmonic behavior of solids.

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## FIGURE CAPTIONS

Fig. 1. The quantity $\varphi_1(T,P)$ defined by Eq. (11) versus the ratio $P/B(T,0)$, for various values of the parameter n.

Fig. 2. The ratio $n/\delta(T,0)$ which satisfies Eq. (15) with the equality sign, versus the ratio $\delta(T,0)/\beta(T,0)$, for various values of the parameter n.

Fig. 3. The quantity $\varphi_2(T,P)$ defined by Eq. (18) versus the ratio $P/B(T,0)$, for various values of the parameter n.

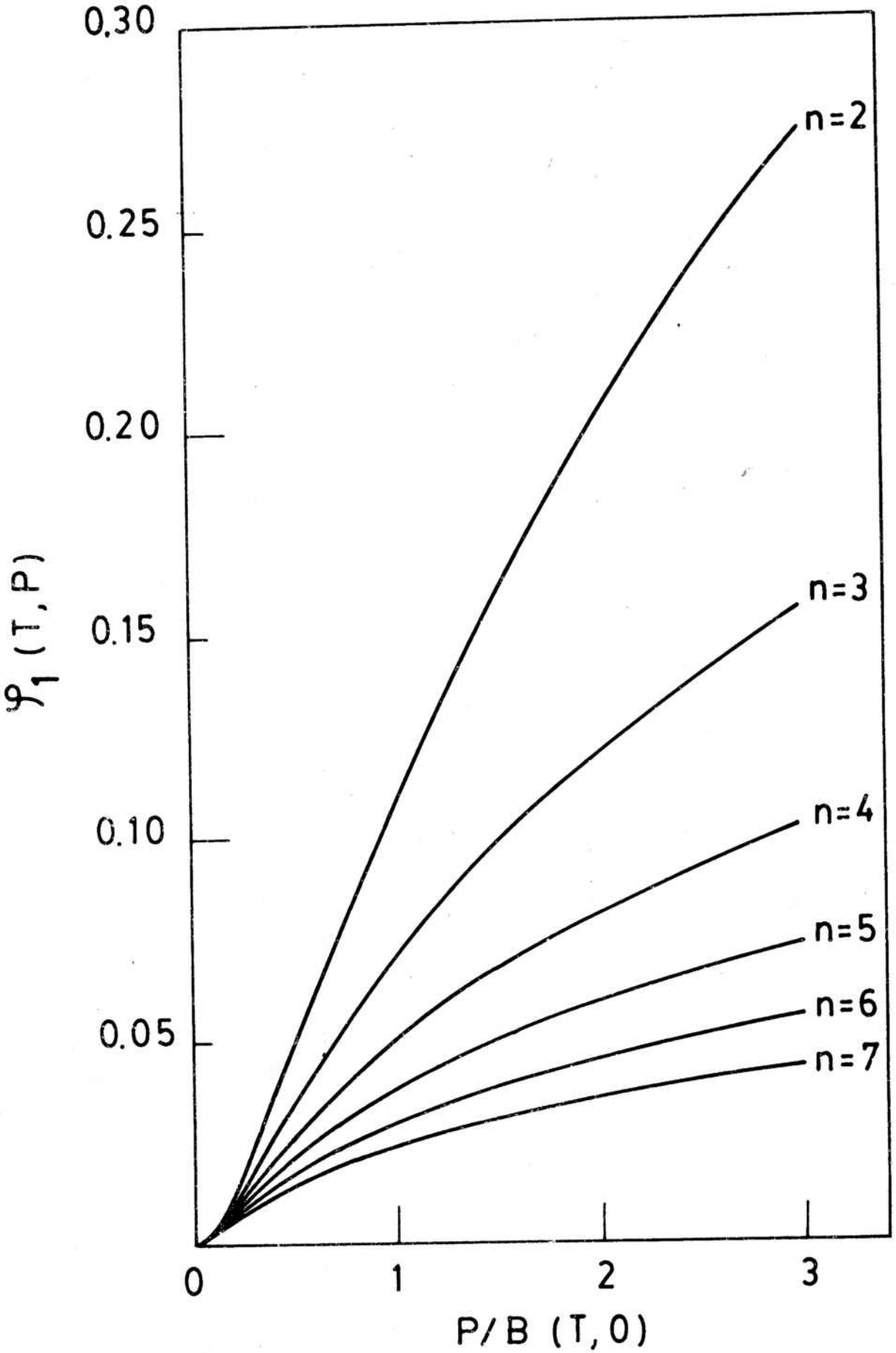


Fig. 1

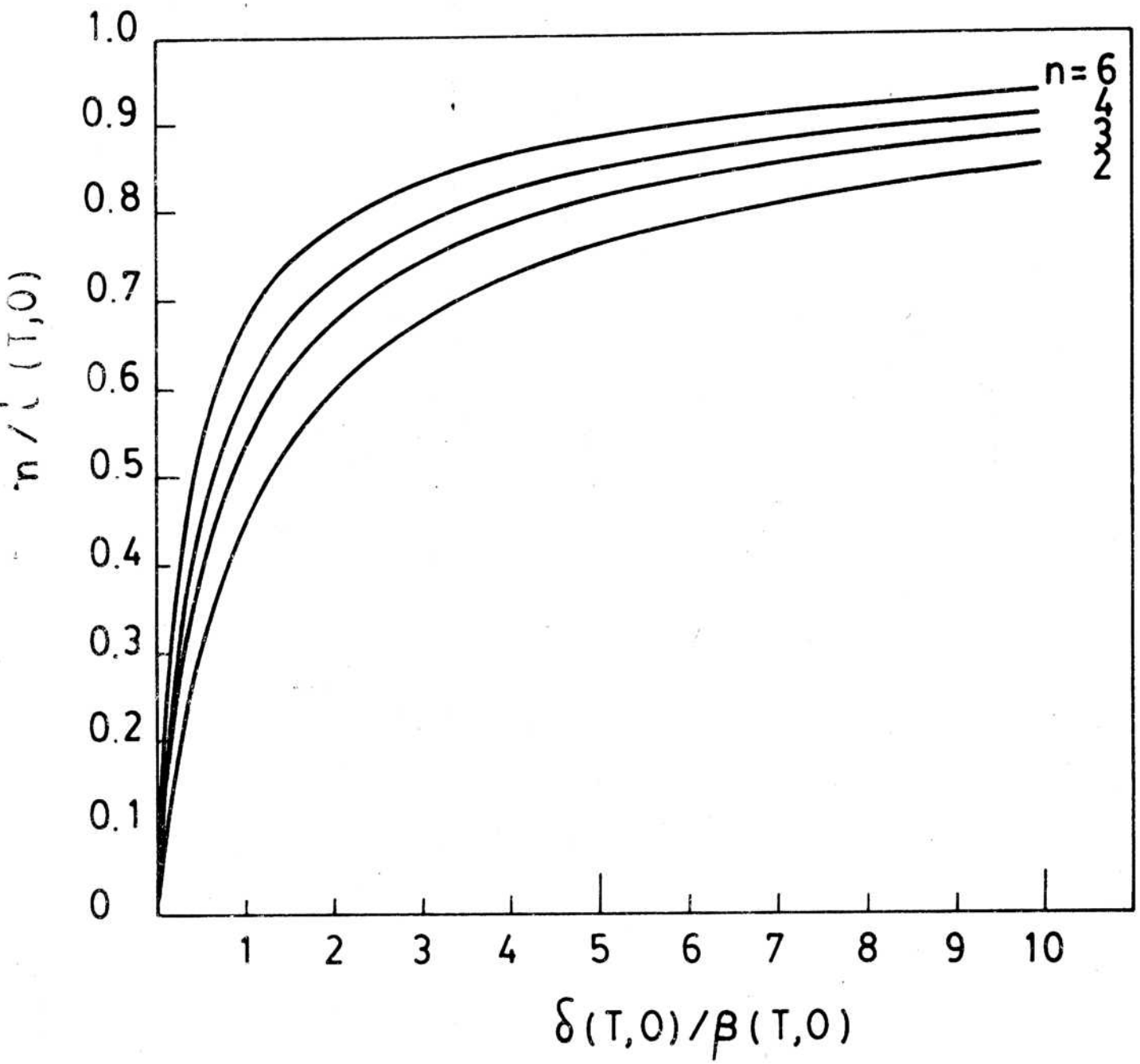


Fig. 2

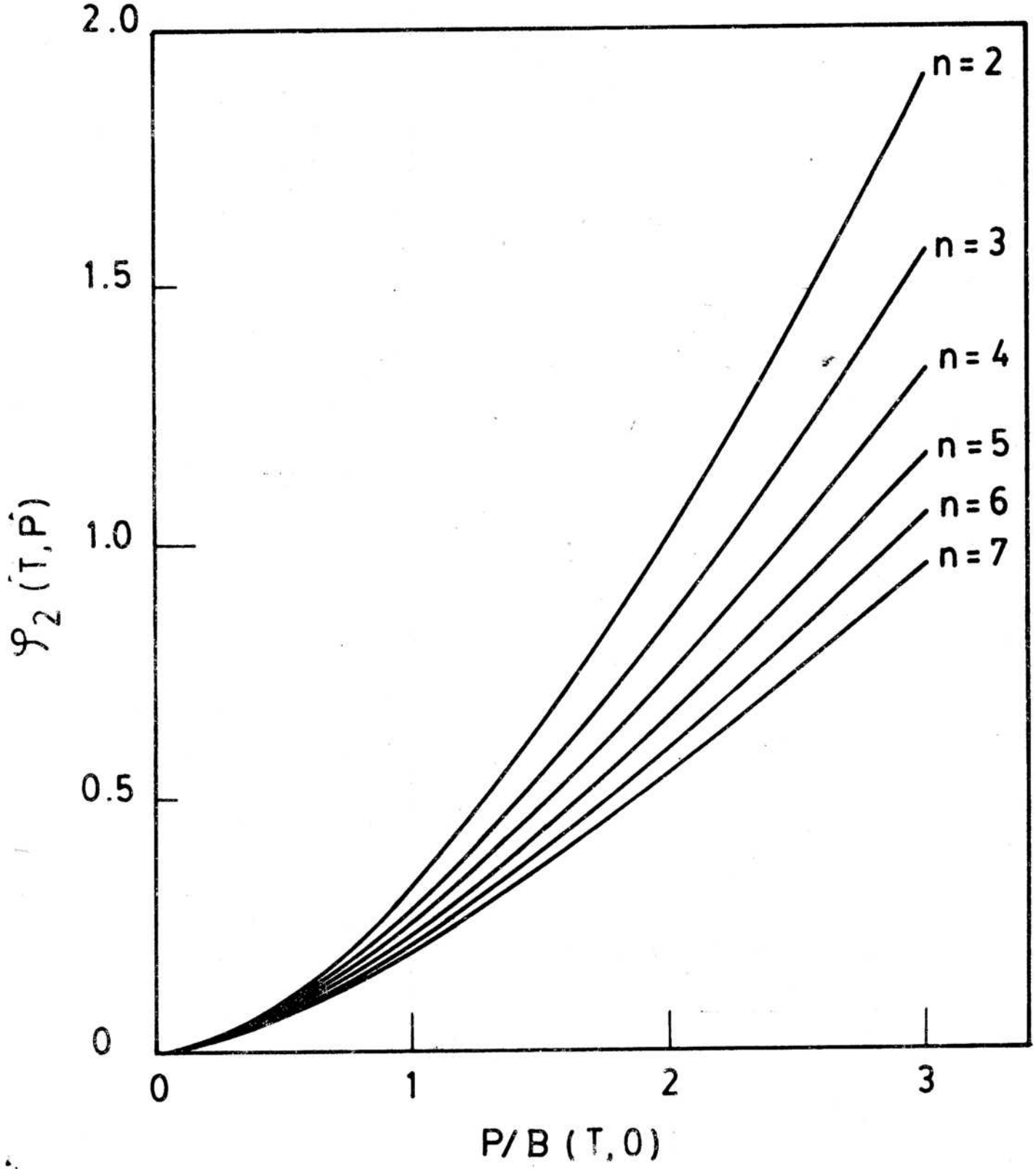


Fig. 3