

# PHYSICAL REVIEW B

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### Universality relationships in condensed matter: Bulk modulus and sound velocity

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We present new forms for the bulk modulus and sound velocity of solids under compression, based on the universal equation of state of Vinet, Ferrante, Smith, and Rose. We then compare the Vinet *et al.* expressions with a number of bulk modulus formulas previously utilized in high-pressure studies. We demonstrate that this form yields a superior fit to experimental data to very high compressions, for a very wide range of solids. These solids cover the entire range of values of the pressure derivative of the bulk modulus which has been observed in high-pressure measurements.

#### INTRODUCTION

Significant progress has been made over the past several years toward the goal of describing the properties of condensed matter in terms of universal relationships involving a small number of parameters. Recent work has demonstrated that the ground-state energy versus atomic separation for a large number of metals obeys a universal relationship of the form

$$E(R_{WS}) = \Delta E E^*(a^*), \quad (1)$$

where  $E^*(a^*)$  is a universal function of  $a^* = (R_{WS} - R_{WS_e})/L$ ,  $\Delta E$  is the energy at equilibrium spacing  $R_{WS_e}$ ,  $L$  is an appropriately defined scaling length, and  $R_{WS}$  is the Wigner-Seitz radius.

Rose, Ferrante, and Smith<sup>1</sup> calculated the adhesive binding energy of all pairs of interfaces formed from Al, Mg, Na, and Zn and found that the binding energy obeyed a universal relation of the above form, where  $L$  was chosen as the average of the Thomas-Fermi screening lengths of the bulk metals. They also found that the bulk energy of Mo, K, Sm, Ba, and Cu was described by a universal energy-distance relationship of the same form. Smith, Ferrante, and Rose then obtained a similar relationship for the chemisorption of H, Li, Na, K, Cs, and Rb atoms on jellium.<sup>2</sup> Next they showed that the binding energy of bulk Mo metal, the diatomic molecule  $H_2^+$ , chemisorbed oxygen, as well as Al-Zn interfaces, exhibited simple two-parameter scaling, and thus could be expressed in terms of a universal binding-energy-distance

relation.<sup>3,4</sup> Guinea, Rose, Smith, and Ferrante<sup>5</sup> differentiated the universal energy relationship with respect to atomic volume  $V = 4\pi(R_{WS})^3/3$  and obtained an equation of state for metals and alloys

$$P(V) = -[\Delta E / (36\pi V^2)L] E'^*(a^*), \quad (2)$$

where a prime indicates differentiation with respect to  $a^*$ . Later they showed that the shape of the force relation,  $F(V) = 4\pi R_{WS}^2 P(V)$ , can be transformed into the universal form<sup>6</sup>

$$F(V) = \Delta F^*(a^*), \quad (3)$$

where  $\Delta F = \Delta E/L$ , and  $F^*(a^*) = -E'^*(a^*)$ . Alternatively, one may define  $G^* = F^*/a^*$ , and thus

$$G^*(a^*) = 4\pi R_{WS}^2 L^2 P(V) / \Delta E (R_{WS} - R_{WS_e}), \quad (4)$$

where  $V_0 = (4\pi/3)R_{WS_e}^3$ ,  $V_0$  is the equilibrium atomic volume or in terms of experimental quantities

$$G^*(a^*) = [X^2/3(X-1)]P(V)/B_0, \quad (5)$$

where  $X = (V/V_0)^{1/3}$  and the scaling length  $L$  for metals is fixed by

$$L = (\Delta E / 12\pi B_0 R_{WS_e})^{1/2}. \quad (6)$$

The universal force function  $F^*(a^*)$  can be related to experimental quantities by the following:

$$F^*(a^*) = (R_{WS_e}/3L)X(V)^2P(V)/B_0. \quad (7)$$

Rose *et al.* scaled experimental pressure-volume data for

Cu, Ag, Li, Rb, Pt, Pd, Mo, and stainless steel.<sup>6</sup> They obtained good agreement with a theoretical estimate for  $F^*(a^*)$ , namely

$$F^*(a^*) = a^*(1 - 0.15a^* + 0.05a^{*2})\exp(-a^*). \quad (8)$$

Now note, if one forms

$$H(V) = -B_0 G^*(a^*) = [X^2/3(1-X)]P(V) \quad (9)$$

and plots  $\ln H$  versus  $(1-X)$ , a straight line is obtained if  $G^*(a^*)$  is a universal function of  $a^*$ . Vinet *et al.*<sup>7</sup> made such plots for a number of different solids under compression and verified that, to a good approximation,  $G^*(a^*)$  is a universal function of  $a^*$  for all classes of solids. Thus they obtained an isothermal equation of state for solids under compression in the absence of phase transitions. Recently Vinet *et al.*<sup>8</sup> extended these results to include temperature effects, and thus were able to predict other isotherms from a reference isotherm, the temperature dependence of the bulk modulus, and the expansion coefficient above the Debye temperature. In addition, Jacobsen and Vinet<sup>9</sup> have applied the analysis to liquid lubricants which undergo solidification transitions at elevated pressures.

Recently Dobson<sup>10</sup> proposed a universal expression for the bulk modulus. Other bulk modulus expressions previously utilized include the well-known Birch-Murnaghan<sup>11</sup> expression, an exponential expression due to Grover, Getting, and Kennedy,<sup>12</sup> the Murnaghan expression,<sup>13</sup> and the Keane<sup>14</sup> expression. All of these forms give a reasonable approximation to the experimental data over some range of compression. In order to elucidate the properties of solids under compression, and to obtain useful expressions for the analysis of experimental data, it is of interest to explore the relationship between the various equations for the bulk modulus.

In this paper we first present two new expressions for the bulk modulus and speed of sound based on the universal relationships of Vinet *et al.*<sup>7</sup> These expressions are demonstrated to be useful for the analysis of experimental data over an extremely large range of compressions. Next we compare the various bulk modulus expressions by means of expansions in powers of the variable  $y = 1 - X$  and demonstrate that all the expressions are equivalent for small compressions. Finally, the various equations are compared with experiment in order to determine the range of validity of the different forms.

#### NEW EXPRESSION FOR THE BULK MODULUS

We observe that a large body of data, obtained in shock-tube experiments over an extremely wide range of compressions, are currently available in the literature.<sup>15,16</sup> Extensive tabulations of density  $\rho$  and sound velocity  $c$  as functions of pressure for room-temperature isotherms exist. These tabulated variables have been extrapolated from the experimentally determined quantities on the Hugoniot curves. We find that these data can be fit by a universal function of density and sound velocity. This has enabled us to predict the behavior of the bulk modulus,  $B$ , for these materials, over a very large range of pressures. Conversely, in the case of solid hydrogen

and deuterium, one may predict the variation of the sound velocity from the bulk modulus measurements over a wide range of pressure.

It was found in previous work<sup>7</sup> that the pressure isotherms for a wide range of materials were very well represented by

$$P(V) = B_0 [3(1-X)/X^2] \exp[\eta(1-X)], \quad (10)$$

$$P(V) = B_0 [3y/(1-y)^2] \exp(\eta y), \quad (10')$$

where  $\eta = \frac{1}{2}[(\partial B/\partial P)_{P=0} - 1]$ , and  $y = (1-X)$ . Thus  $\ln H = \ln B_0 + \eta y$  and the  $\ln H$  versus  $y$  plots are straight lines of slope  $\eta$  and intercept  $\ln B_0$ . The isothermal bulk modulus is given by

$$B = (B_0/X^2)[2 + (\eta - 1)X - \eta X^2] \exp[\eta(1-X)], \quad (11)$$

$$B = [B_0/(1-y)^2][1 + (\eta + 1)y - \eta y^2] \exp(\eta y). \quad (11')$$

We proceed by forming  $b(X) = X^2 B(V)$ . Thus

$$b(X) = B_0 [2 + (\eta - 1)X - \eta X^2] \exp \eta(1-X) \quad (12)$$

or

$$b(y) = B_0 [1 + (\eta + 1)y + \eta y^2] \exp \eta y. \quad (12')$$

Therefore,

$$\ln b = \ln B_0 + \eta y + \ln [1 + (\eta + 1)y - \eta y^2].$$

The adiabatic bulk modulus is related to the sound velocity and density by

$$B_A = \rho c^2 = \rho_0 c^2 / X^3. \quad (13)$$

The isothermal bulk modulus

$$B_T = (C_V / C_P) B_A,$$

where the  $C$ 's are the specific heats at constant pressure and constant volume, respectively. Thus

$$\ln b_A = \ln(C_P / C_V) + \ln b_T = (2 \ln v - \ln X), \quad (13')$$

where  $v = \rho_0^{1/2} c$ .

In Fig. 1 we present plots of  $\ln b_A$  versus  $y$  for several solids except for  $H_2$ , where we have used  $b_T$ . In Fig. 2 we present plots of  $\ln v$  versus  $y$  for the same solids. The solids chosen cover the entire range of  $\eta$ 's characteristic of the materials which have been investigated experimentally. We observe that these plots are quite linear over a considerable range of  $y$ . We have shown that this result would be essentially true, whether using the isothermal or adiabatic bulk modulus. A direct comparison of the two using the speed of sound to calculate  $B_A$  and derivatives of the isothermal  $P$ - $V$  curves for  $B_T$  gave a maximum deviation in  $\ln b$  of 5% and a mean deviation of  $(0.15 \pm 0.13)\%$  for the 19 materials for which we had data.<sup>15</sup>

In our analysis of the experimental data, we proceeded by calculating  $\ln v$  and  $\ln H$  from the tabulated values of  $c$ ,  $\rho$ , and  $P$ . Then we obtained  $\ln b$  from  $\ln v$ , using Eq. (13'), rather than from  $\ln H$ . This has the advantage that one need not input an  $\eta$  value. The values of the zero-pressure bulk modulus  $B_0$  obtained from least-square fits

of the  $\ln b$  and  $\ln H$  plots, are in good agreement with each other.

In the case of solid hydrogen and deuterium, one has available values of  $V/V_0$ ,  $P$ , and  $B$ . Hence one may utilize Eq. (13') to obtain  $\ln v$ . Thus we have presented an alternate method for analyzing experimental bulk modulus and sound velocity data for solids under compression. We note that the  $\ln b$  and  $\ln v$  plots are, to a very good approximation, linear in  $y$  for materials in which there are no phase transitions over the observed pressure range.

**COMPARISON WITH OTHER BULK MODULUS FORMULAS**

We now proceed to compare several previously utilized expressions for the bulk modulus of solids, under compression and compare them to Eq. (11'). The expressions due to Dodson,<sup>10</sup> Birch,<sup>11</sup> Grover *et al.*,<sup>12</sup> Murnaghan,<sup>13</sup> and Keane<sup>14</sup> will be shown to have identical leading terms for small compressions, as Eq. (11').

Expanding  $\exp(\eta y)$  in powers of  $y$ , Eq. (11') for the bulk modulus becomes

$$B = B_0 [1 + (2\eta + 1)y + 3\eta y^2/2 + O(y^3)] / (1 - y)^2 \quad (14)$$

Now we expand the term in  $(1 - y)^2$  to obtain an alternate expression for  $B$ ,

$$B = B_0 \{ 1 + (2\eta + 3)y + [\frac{3}{2}\eta^2 + 4\eta + 3]y^2 + O(y^3) \} \quad (14')$$

Grover, Getting, and Kennedy<sup>12</sup> utilized the following expression for  $B$ :

$$B = B_0 \exp(\alpha \Delta V / V_0) \quad (15)$$

which may be written in terms of  $X$  as

$$B = B_0 \exp[\alpha(1 - X^3)] \quad (16)$$

Expanding the exponential in powers of  $(1 - X^3)$ , we obtain

$$B = B_0 [1 + \alpha + \alpha^2/2 - \alpha(1 - \alpha)X^3 + \alpha X^6/2 + \dots] \quad (17)$$

Equation (21) reduces to the following when  $X^3$  and  $X^6$  are expanded in  $y$ :

$$B + B_0 [1 + 3\alpha y - 3\alpha(1 - \alpha)y^2 + O(y^3)] \quad (18)$$

Note that the leading terms of this expression for  $B$  are identical to those of Eq. (14) if one equates  $3\alpha$  to  $(2\eta + 3)$ , where  $\eta = \frac{3}{2}(B'_0 - 1)$ . Thus we find that

$$\alpha = B'_0 \quad (19)$$

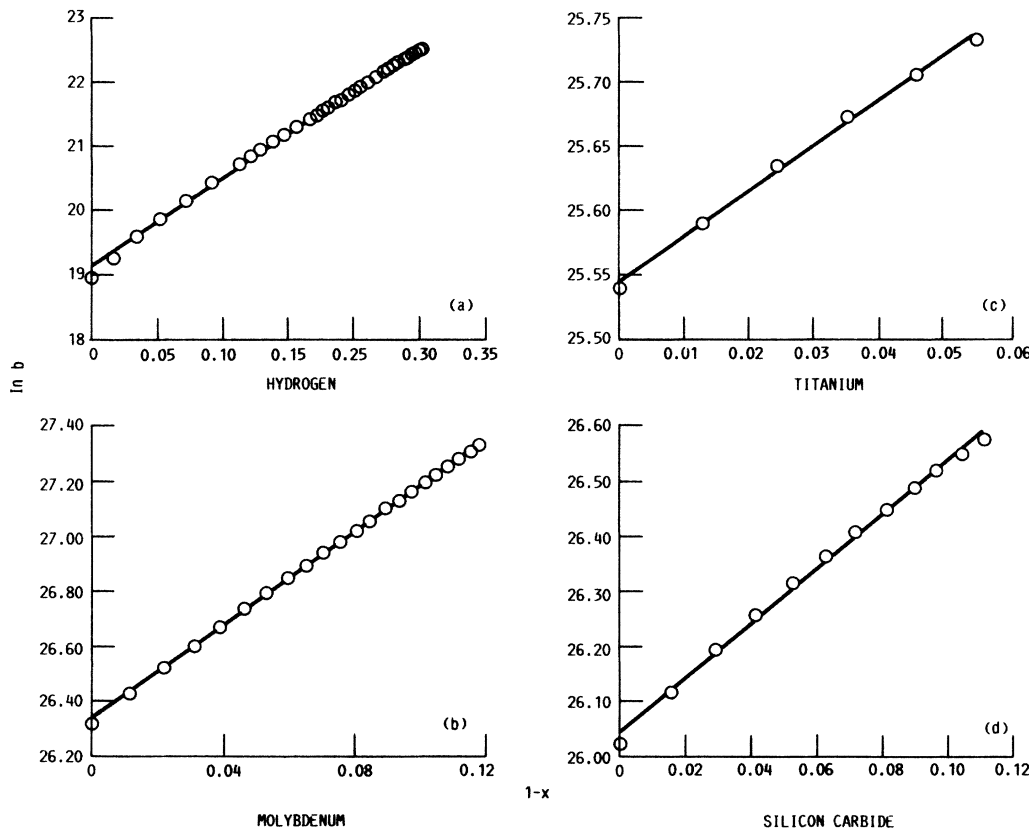


FIG. 1. A plot of  $\ln b [b = X^2 B(X)]$ , vs  $(1 - X) [X = (V/V_0)^{1/3}]$ , for (a) hydrogen (Refs. 17 and 18), (b) molybdenum (Ref. 15), (c) titanium (Ref. 15), and (d) silicon carbide (Ref. 15).

Next we show that the expansion of the following form for  $B$  due to Dodson<sup>10</sup> has the same leading terms, for small  $y$ , as our expression

$$B = \alpha[(1/X) - \beta]^2. \quad (20)$$

where  $\alpha = B_0(1-\beta)^{-2}$  and  $\beta = 1 - (2/3B'_0)$ . Thus  $\alpha = 9B_0B_0'^2/4$  and

$$B = (9B_0B_0'^2/4)[(1/X) - 1 + (2/3B'_0)]^2. \quad (21)$$

Substituting  $y = 1 - X$ , we obtain, after some algebraic manipulation

$$B = B_0[1 + (2\eta + 1)y + (2\eta + 1)y^2/4]/(1-y)^2. \quad (22)$$

Thus Dodson's form for  $B$  has the same leading terms, for small  $y$ , as Eq. (14).

Another equation of state due to Birch,<sup>11</sup> which has been extensively utilized in geophysics and is based on an expansion of the elastic strain energy to second order, may be written as

$$P = 3B_0(1-X^2)[1 + (\eta/2 - \frac{9}{4})(1-X^2)/X^2]/2X^7. \quad (23)$$

This yields the following form for  $B$ , which has the same leading terms as Eq. (14):

$$B = B_0X^{-9}[9w + 7(1-2w)X^2 + 5(w-1)X^4]/2, \quad (24)$$

where  $w = \eta/2 - \frac{9}{4}$ . Expanding in powers of  $y$ , we obtain

$$B = B_0[1 + (4w + 10)y + (64w - 55)y^2/2 + O(y^3)]/(1-y)^2 \quad (25)$$

or in terms of  $\eta$

$$B = B_0[1 + (2\eta + 1)y + (32\eta - 199)y^2/2 + O(y^3)]/(1-y)^2. \quad (26)$$

Another bulk modulus formula, used in geophysics, which has the same leading terms as Eq. (14) is that of Keane.<sup>14</sup> It may be written as

$$B = B_0[1 + (2\eta + 3)(X^{-5} - 1)/5], \quad (27)$$

which, upon expansion of  $X^{-5}$  in powers of  $y$ , reduces to

$$B = B_0[1 + (2\eta + 3)y + (4\eta + 6)y^2 + O(y^3)]. \quad (28)$$

The Murnaghan<sup>13</sup> bulk modulus formula is given by

$$B = B_0(V_0/V)^{B'_0} = B_0(1-y)^{-(2\eta+3)} \quad (29)$$

whose expansion in powers of  $y$  is

$$B = B_0[1 + (2\eta + 3)y + (2\eta^2 + 7\eta + 6)y^2 + O(y^3)]. \quad (30)$$

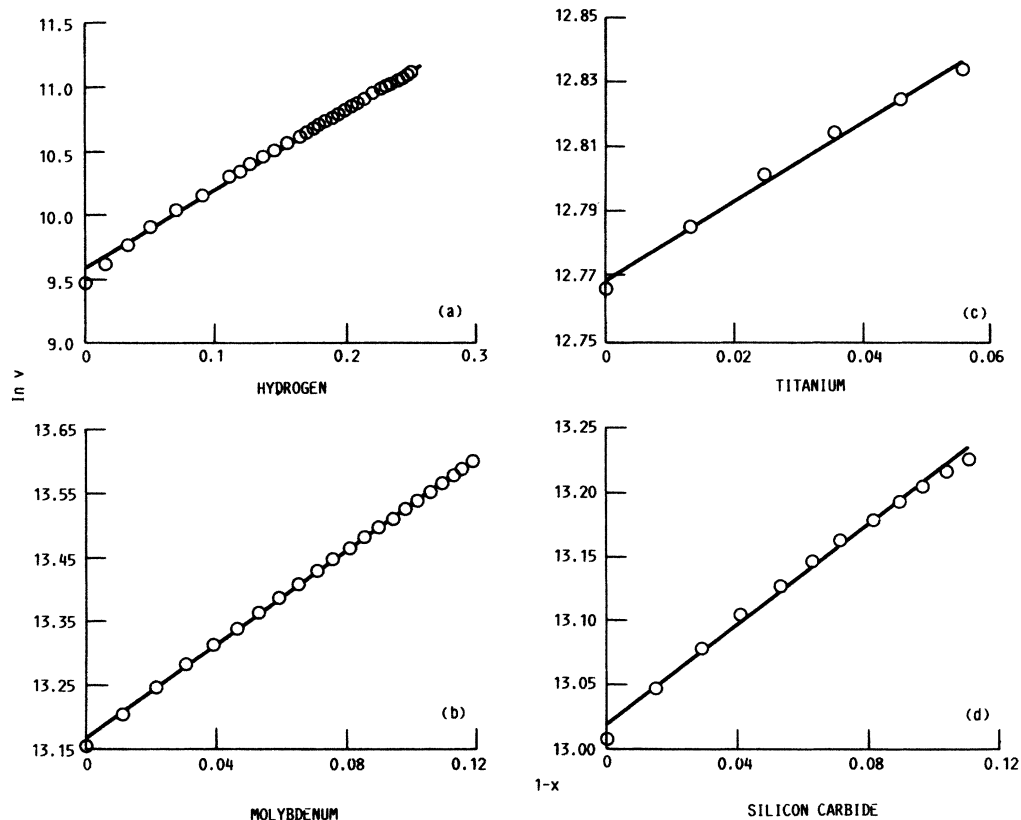


FIG. 2. A plot of  $\ln v$  ( $v = \rho^{1/2}c$ ), vs  $(1-X)$  [ $X = (V/V_0)^{1/3}$ ], for (a) hydrogen (Ref. 17), (b) molybdenum (Ref. 15), (c) titanium (Ref. 15), and (d) silicon carbide (Ref. 15).

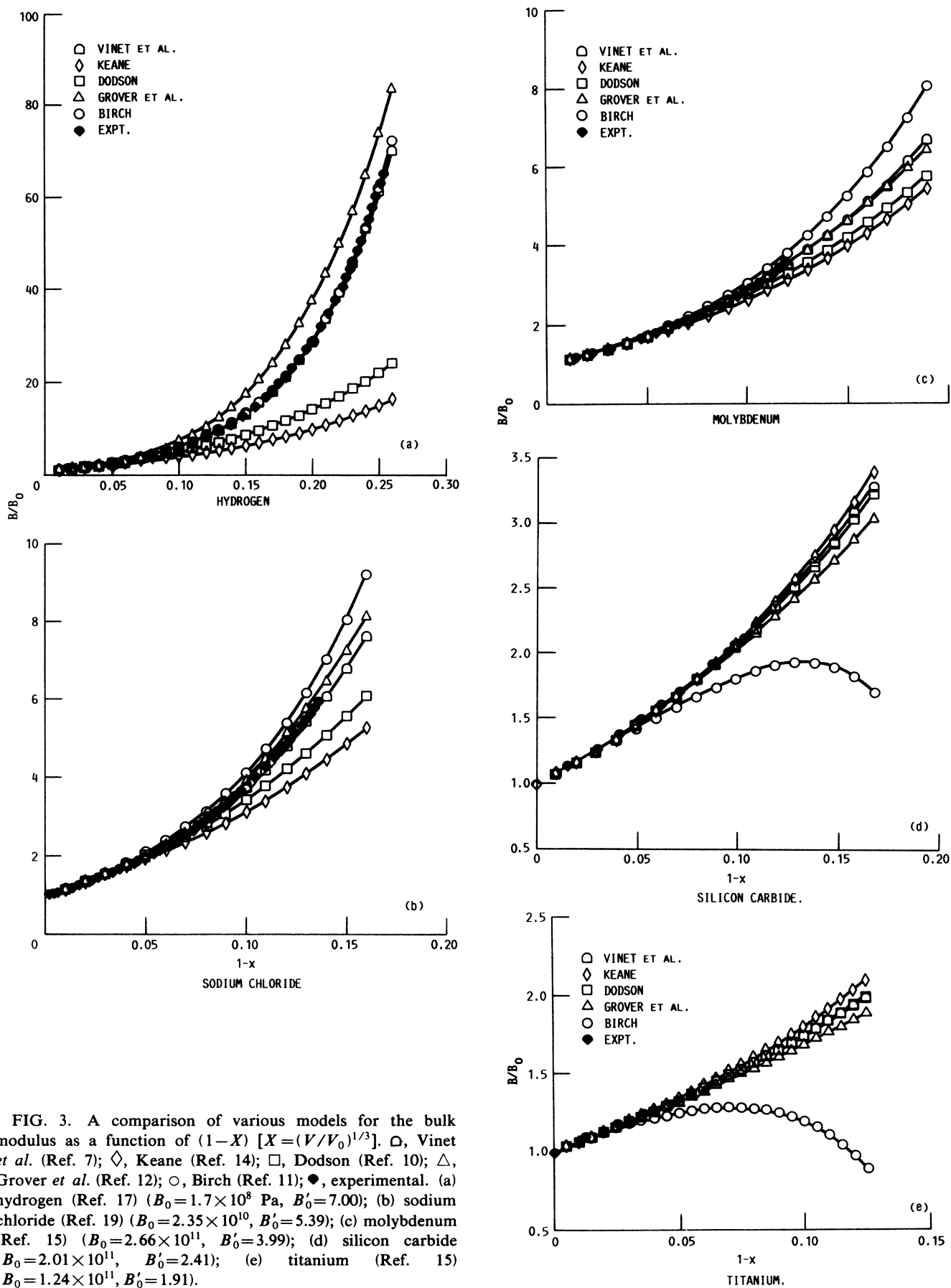


FIG. 3. A comparison of various models for the bulk modulus as a function of  $(1-X)$  [ $X=(V/V_0)^{1/3}$ ].  $\square$ , Vinet *et al.* (Ref. 7);  $\diamond$ , Keane (Ref. 14);  $\square$ , Dodson (Ref. 10);  $\triangle$ , Grover *et al.* (Ref. 12);  $\circ$ , Birch (Ref. 11);  $\bullet$ , experimental. (a) hydrogen (Ref. 17) ( $B_0=1.7 \times 10^8$  Pa,  $B'_0=7.00$ ); (b) sodium chloride (Ref. 19) ( $B_0=2.35 \times 10^{10}$ ,  $B'_0=5.39$ ); (c) molybdenum (Ref. 15) ( $B_0=2.66 \times 10^{11}$ ,  $B'_0=3.99$ ); (d) silicon carbide ( $B_0=2.01 \times 10^{11}$ ,  $B'_0=2.41$ ); (e) titanium (Ref. 15) ( $B_0=1.24 \times 10^{11}$ ,  $B'_0=1.91$ ).

The Murnaghan expression is a first-order expansion of the elastic strain energy and is thus linear in the pressure. Therefore, it is not expected to represent the nonlinear terms arising at high compressions.<sup>5,14</sup>

Thus the various expressions for the isothermal bulk modulus which we have considered all have the same leading terms for small compressions (small  $y$ ).

#### COMPARISON OF BULK MODULUS EXPRESSIONS WITH EXPERIMENT

We now compare the various bulk modulus expressions with each other and with experimental data for a range of  $\eta$  values. In Figs. 3(a)–3(e) the various expressions are plotted for  $\eta$ , varying from 1.36 to 9. We observe that the curves all agree with each other for small  $y$  (low compression). This is understandable since the leading terms of the expansions of all the bulk modulus expressions, in powers of  $y$ , are identical. However, at high compressions, substantial differences exist between the various forms of all values of  $\eta$ . For solids with large  $\eta(B'_0)$  values, the formulas of Vinet *et al.* and Birch fit the experimental data most accurately over the entire range of  $y$ . For smaller  $\eta$ , the expressions of Vinet *et al.* and Grover *et al.* yield the most accurate representation of the data. Several different methods were utilized for selecting the values of  $B_0$  and  $B'_0$  used in the comparisons. For NaCl we used the independent ultrasonic values of  $B_0$  and  $B'_0$ . For hydrogen we used the values reported by Anderson and Swenson, and for Ti and Mo we used values obtained from  $\ln H$  versus  $y$  plots.

Since there is some ambiguity in selecting values of  $B_0$  and  $B'_0$  from compression data, we have performed a further comparison which is model independent. The hydrogen bulk modulus data of Anderson and Swenson is given for a very wide range of compressions ( $X=0.4-1$ ). Thus the ability to fit this data accurately is a severe test of each model. We have performed a least-squares fit to the hydrogen data for each of the functions plotted. The results are given in Table I as  $Q = 1 - \sum[(\text{rms deviation})/(\text{experimental value})]$ . Where possible we have used the expressions for the pressure for each model to fit the data, since the bulk modulus data is

TABLE I. A comparison of  $B_0$  and  $B'_0$  for least-squares curve fits to the Anderson and Swenson hydrogen data between the various theoretical expressions.

	$B_0$ , Pa	$B'_0$	$Q$
Vinet <i>et al.</i> <sup>a</sup>	$1.62 \times 10^8$	7.45	0.9725
Grover <i>et al.</i> <sup>b</sup>	$9.65 \times 10^7$	8.02	0.8811
Keane <sup>c</sup>	$-1.88 \times 10^9$	-3.1	0.2535
Birch <sup>d</sup>	$1.7 \times 10^8$	7.23	0.9999
Dodson <sup>e</sup>	$7.77 \times 10^5$	210.65	1.765

<sup>a</sup>Reference 7.

<sup>b</sup>Reference 12.

<sup>c</sup>Reference 14.

<sup>d</sup>Reference 11.

<sup>e</sup>Reference 10.

obtained by taking derivatives of pressure data. The present model and the Birch formula give the best fit to the data over this wide compression range. Since Anderson and Swenson used the Birch equation to smooth their experimental data, it is not surprising that it fits the hydrogen data so well. However, as can be seen for Ti, Fig.

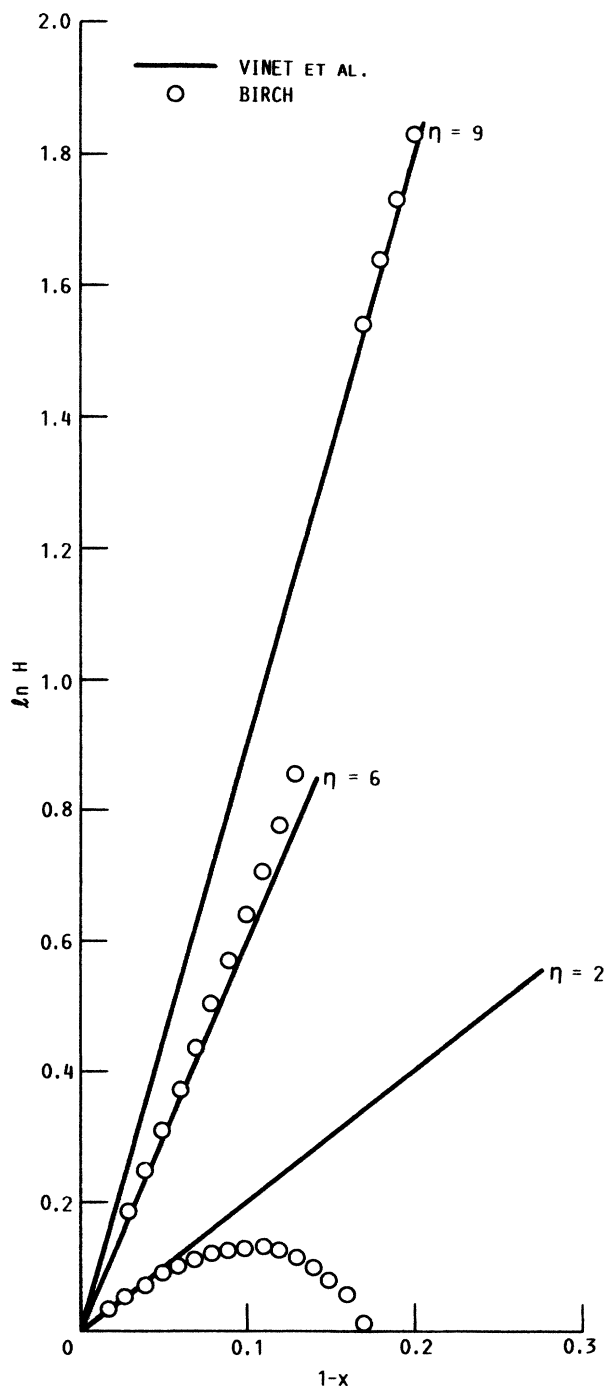


FIG. 4. A comparison of  $\ln H$  ( $H = PX^2/[3(1-X)]$ ), vs  $(1-X)$  [ $X = (V/V_0)^{1/3}$ ] between the Vinet *et al.* (Ref. 7) expression for the pressure, and the Birch equation (Ref. 11) for the pressure for a range of values of  $\eta(B'_0)$  representative of experimental data.

3(e), the Birch equation fails at large compressions when  $\eta$  is small (1.37). Examination of Eq. (23) shows that the  $X$  dependence reverses sign for  $\eta=4.5$  ( $B'_0=4$ ). This yields a nonphysical decrease of the bulk modulus with increasing  $y$ , without a phase change, for  $\eta < 4.5$ . Thus we may conclude that the present expression gives the best representation of the experimental compression data over the widest range of parameters used ( $B_0, B'_0, V/V_0$ ).

Since the Birch equation is extensively used at present for analysis of high-pressure data, we make a further comparison. In Fig. 4 we plot  $\ln H$  versus  $y$  for  $\eta$  varying between 2 and 9 for both the Vinet *et al.* model and the Birch equation. We can see that for large  $\eta$  the Birch equation and the Vinet *et al.* model yield essentially the same functional form. Without further experimental data, at large compressions, for materials with large  $\eta$ , it is difficult to choose between them. However, our equation has the advantage of representing the widest range of experimental data and giving a particularly simple method for data analysis utilizing the  $\ln b$  and  $\ln v$  versus  $y$  plots.

There is one final point to be made concerning data analysis and the fitting of experimental data. Each of the bulk modulus formulas discussed have  $B_0$  and  $B'_0$  appearing explicitly. The values of  $B_0$  and  $B'_0$  obtained by fitting the experimental data may differ significantly from the values given by fitting the small compression experimental data. We have found that the values of  $B_0$  and  $B'_0$  obtained from the Vinet *et al.* model are relatively insensitive to the number of experimental data points used in the fitting procedure; all the other expressions are

much more sensitive to the number of data points utilized in the fit. A comparison of the relative sensitivity of the various expressions is also given in Table I. For example, for hydrogen the Murnaghan equation yields values for  $B_0$  and  $B'_0$  of  $2.68 \times 10^8$  Pa and 4.26 with  $Q=0.92$  using all of the data, compared to values of  $1.7 \times 10^8$  Pa and 7 obtained using the small compression data.

In conclusion, we have presented several new, generally valid expressions for the analysis of compression data and determination of the bulk modulus of solids under high compression. Expressions widely used in the literature have been expanded in terms of the compression and compared with each other. All of the expressions agree with each other to first order; however, they differ appreciably at high compressions. Compression data for solids having a wide range of  $\eta$  ( $B'_0$ ) values has been analyzed. The  $\eta$  values of the solids studied is characteristic of the entire range of available experimental data. We have found that the EOS of Vinet *et al.* yields the best representation of the experimental data over the entire range of parameter values ( $B_0, B'_0, V/V_0$ ) thus far observed in condensed matter under compression.

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