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An Equation of State for Metals

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Abstract

A new analytical equation of state is offered for metals. The form is expressible completely in terms of the maximum lattice vibration frequency, macroscopically expressible as the characteristic temperature, Θ, and its derivatives. For one particularly useful special case, the results have been found to match both cold-curve and shock-Hugoniot data for a variety of metals. The equation is of the Grüneisen variety, in which the Grüneisen parameter is specified as a function of volume only. The equation begins to lose accuracy in the pressure range of one to several megabars, perhaps because of the breakdown of the Grüneisen assumption, whereupon the Grüneisen parameter becomes energy dependent.

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1. INTRODUCTION

An analytical equation of state, satisfying the Grüneisen assumption, was developed for metals, which matches available cold-curve and Hugoniot data for a variety of metals out to pressures in the megabar regime. The model uses the Universal Cold Curve (UCC) model of Rose *et al.* [1] as its inspiration, rather than being derived from first principles. The UCC model matches cold-compression data for a variety of metallic elements, over wide ranges of pressure into the megabar range. However, unlike the UCC model, which only gives pressure along the zero-degree isotherm (*i.e.*, the cold curve) in terms of a density variable, the currently offered model is expressible in terms of higher-level thermodynamic functions (namely, the maximum lattice vibrational frequency, ω_{max}, macroscopically expressible as the characteristic temperature, $\Theta(V)$, and its derivatives). The current model therefore allows the calculation of state changes along any thermodynamic path, not only that of isothermal, cold compression. Unlike other follow-on models to Rose *et al.*, such as Vinet *et al.* [2, 3] and Baonza *et al.* [4, 5], the current model's thermal component is coupled to the lattice compression component in a unified way, precisely because the lattice compression component is expressed in terms of frequency, not compression. The general form of the current model requires the specification of the characteristic temperature (*i.e.*, lattice frequency) with volume; however, one particular form for $\Theta(V)$ has been employed, with very good resulting agreement to both cold-curve and shock-Hugoniot data for metals. As a note of warning though, the use of the model for materials undergoing phase change has not been validated, and is not therefore encouraged.

2. BACKGROUND

The UCC model [1] does a truly remarkable job of matching cold-compression data for a variety of solid elements, over wide ranges of pressure into the megabar range. The

UCC model also predicts the tensile pressure in materials under cold expansion out to rather large distentions. The UCC model expresses the cold energy as

$$E_c = E_b\left[1 - (1 + a + 0.05a^3)\exp(-a)\right] \;, \tag{1}$$

where E_b is the specific lattice binding energy, and a is a dimensionless parameter defining the relative spacing of the lattice atoms, given by

$$a = \eta\left((V/V_0)^{1/3} - 1\right) \;. \tag{2}$$

In eqn. (2), the parameter η is called the anharmonicity factor and V and V_0 are the current and ambient specific volumes of the material, respectively. The anharmonicity factor represents a ratio of lengths on the atomic scale, but may be expressed in terms of macroscopic parameters as

$$\eta = 3\frac{C_0}{\sqrt{E_b}} \;, \tag{3}$$

where C_0 is the bulk sound speed in the material at zero pressure and absolute temperature. Note that the use of the adiabatic, bulk sound speed is justified, even though Rose *et al.* [1] express their result in terms of the *isothermal* bulk modulus, since the zero-degree isotherm is also an isentrope.

Since the cold curve is not only an isotherm but also an isentrope, the cold pressure, p_c, is obtainable simply as the negative of the derivative of the cold energy ($-dE_c/dV$). The UCC model is essentially a well-reasoned empirical fit to data derived from first-principle models of cohesive energies expressed as a function of lattice spacing. By clever construct, Rose *et al.* [1] developed the nondimensional parameter a, which effectively scales the cold curve for all solids onto the same metric. When applied to available macroscopic cold-compression data for a variety of materials, the model provides an extremely accurate description of available data, encompassing pressures out to one and, in some cases, several megabars.

Since the publication of the the UCC model by Rose *et al.* [1], there have been further advances following the lines of their work. Vinet *et al.* [2, 3] successfully introduced temperature effects into the context of the earlier work. It was shown that the form of Rose was valid, not only for zero temperature, but for any isotherm. The model of Vinet thus became an isothermal equation of state, and temperature effects were accounted for by modeling the variation of bulk modulus with temperature. Though not explicitly casting their work in Grüneisen form, they do so implicitly by citing that "it has been observed experimentally that above the Debye temperature, [thermal pressure] is a linear function of T." By adding to this observation that above the Debye temperature, thermal energy is likewise linear in T, one may conclude that $V(\partial p/\partial E)_V = \Gamma$ (the Grüneisen parameter) is thus constant for fixed V. In both works of Vinet *et al.*, the temperature dependence of the bulk modulus was linearly fit to data for several substances. With this linear fitting form, they showed that their equation of state could reproduce the Murnaghan equation of state.

Very recent works by Baonza *et al.* [4, 5] have been able to advance the field by expressing the compressibility of dense phases using an inverse power law as a function of the divergence pressure along a certain pseudospinodal curve at the considered temperature.

To obtain a general equation of state at points off the pseudospinodal, they too, like their predecessor Vinet, model the thermal pressure deviations from this reference curve using a Grüneisen law, though in their case, it is one in which the Grüneisen law is assumed to follow a commonly used form—namely, (Γ/V) = fitted constant.

A key distinction, however, between the work of Vinet *et al.* [2, 3], Baonza *et al.* [4, 5] and what will be proposed in the current work is that, in the works of Vinet and Baonza, the function governing the Grüneisen reference curve behavior (an isotherm in the case of Vinet, the zero pseudospinodal curve in the case of Baonza) and that governing temperature (*i.e.*, thermal pressure) effects are functionally independent. There is no unifying precept tying the two functions together. By contrast, the current work will predict volumetric and thermal effects with a single unified function. Volumetric effects are defined through the characteristic temperature function, Θ, and thermal effects (via internal energy, E) are defined via the ψ function (related to the Grüneisen function by $\psi=V/\Gamma$), and are thus inextricably linked to Θ by definition. In essence, specification of the cold curve in the context of the current model defines the thermal behavior, or alternately, specification of the Grüneisen function for thermal behavior defines the cold curve. The key to this advance over prior work is modeling the cold curve, not in terms of density, as do Vinet and Baonza, but doing so in terms of lattice frequency, as did Debye and Einstein.

3. THE GRÜNEISEN ASSUMPTION

The characteristic temperature, Θ, is an important thermodynamic parameter. The best model existing to date for it is the Debye temperature, Θ_D. The characteristic temperature is central to the question of how the constant-volume specific heat varies with temperature, from the high temperature limit of three times the universal gas constant per mole (known as the law of Dulong and Petit [6]) to a value of zero at absolute zero temperature. The derivation of specific heat is based on quantum statistical mechanics and depends on the frequency distribution of vibration modes, ω_i, within the crystal lattice. In 1907, Einstein proposed that all permissable vibrational modes are simultaneously excited with temperature, and he derived a specific heat expression that was qualitatively correct, but which approached zero in too rapid of a manner at absolute zero temperature. Debye remedied this deficiency in 1912 by proposing a more realistic frequency distribution of vibrational modes [6]. Common to all of these models is the notion of a characteristic temperature, by which the physical temperature, T, may be scaled in order to achieve the dimensionless temperature parameter, (Θ/T), through which the specific heat is expressed. In all of these models, including that of Debye, the characteristic temperature is directly proportional to the maximum excited frequency mode, ω_{max}.

As Desloge [7] points out though, the characteristic temperature is itself a function of volume [*i.e.*, $\Theta = \Theta(V)$]. The variation of Θ with V is really a macroscopic manifestation of the variation of vibrational frequencies ω_i with V. But since specific heat theory indicates that "the frequency spectrum is entirely determined by the limiting frequency ω_{max} . . . all other oscillations change their frequencies in the same ratio [as ω_{max}] [8]." Thus, the log-derivative of any arbitrary vibrational mode with volume is the same as the log-derivative of the limiting frequency, ω_{max}, namely

$$-\frac{d\ln\omega_i}{d\ln V} = -\frac{d\ln\omega_{\max}}{d\ln V} \quad . \tag{4}$$

This condition quantifies the Grüneisen assumption. Not unexpectedly, its value is known as the Grüneisen parameter, denoted by

$$\Gamma = -\frac{d\ln\omega_{\max}}{d\ln V} \quad . \tag{5}$$

Of course, it is more common to see this parameter expressed in terms of macroscopic thermodynamic state variables as

$$\Gamma = V(\partial p/\partial E)_V \quad . \tag{6}$$

The notion that all vibrational frequencies change with volume in the same ratio as the limiting frequency is fundamental to Grüneisen theory. The alternative is that each vibrational mode would have its own associated Grüneisen parameter, Γ_i, and the aggregate macroscopic effect would necessarily be an average over all of the individual Γ_i, weighted in some manner by the fractional energy component associated with each mode. Such an aggregate Γ would not only vary with volume, but also with thermal energy, as the fractional energy component associated with each mode was redistributed with increasing temperature. In fact, Grodzka [9] summarizes work that suggests there is a significant decrease in the Grüneisen parameter with thermal energy. Similarly, very high pressure equation-of-state theories, which include electronic effects, are clear departures from Grüneisen theory.

Since, however, non-Grüneisen theory is beyond the scope of the current effort, we limit the discussion to materials obeying the constraint of eqn. (4). Since both the characteristic temperature and the Grüneisen parameter are related to the limiting vibration frequency of the lattice, ω_{max} (Θ directly proportional to ω_{max} and Γ related through eqn. [5]), Θ and Γ are, in fact, related to each other as follows:

$$\frac{\Theta'}{\Theta} = -\frac{1}{\psi} \quad , \tag{7}$$

where the prime denotes differentiation with respect to specific volume, and the state variable ψ, defined as V/Γ, has been introduced [10, 11] for convenience in manipulating the thermodynamic equations. The functional form of the Grüneisen parameter will define the quantitative relationship expressed in eqn (7). For example, if Γ is a constant, independent of volume, then

$$\Theta\, V^{\Gamma_0} = \text{constant} \qquad\qquad (\text{when } \Gamma \text{ constant}).$$

If, on the other hand, ψ is held constant [*i.e.*, $\Gamma = \Gamma_0/(1+\mu)$, where $\mu = (V_0 - V)/V$], then

$$\Theta e^{\Gamma} = \text{constant} \qquad\qquad (\text{when } \psi \text{ constant}).$$

Finally, if ψ is linear in V, such that ψ' is held constant [*i.e.*, $\Gamma = \Gamma_0/(1+\beta\mu)$], as Segletes [12] proposed for reasons of thermodynamic stability, then

$$\Theta\, \psi^{(1/\psi')} = \text{constant} \qquad\qquad (\text{when } \psi' \text{ constant}).$$

This last Grüneisen form is equivalent to that known as the Sesame formula [13]. Each unique expression for Γ will produce a unique algebraic relationship between Γ and Θ, though each will satisfy the differential relationship of eqn. (7).

The Grüneisen equation of state, which follows from the Grüneisen assumption, may be expressed in a variety of ways, but amounts to the following:

$$p - p_{ref} = (E - E_{ref})/\psi \quad . \tag{8}$$

In this equation, the reference functions are the pressure and specific internal energy states along a known reference curve, such as an isotherm, isentrope, or Hugoniot. One of the historical problems with estimating the Grüneisen function has been that it was usually inferred by using hard experimental data along one thermodynamic path (*e.g.*, the Hugoniot) and comparing it to *predictions* of thermodynamic states along another path [14–16] (usually the cold curve predicted by models of Slater [8], Dugdale and McDonald [17], or others). In this sense, previous nonambient Grüneisen values were not even obtained by indirect measurement, but rather were back-calculated by assuming the validity of a particular cold-curve model. Grodzka [9] cleverly displayed the result of using this inference approach for Grüneisen estimation, by showing widely varying estimations of the Grüneisen function for aluminum, depending upon whose data and whose model was employed. Segletes [10–12] has investigated the thermodynamic stability of the Grüneisen equation and has been able to place some constraints upon allowable functional behavior of the Grüneisen function. As modeling and measurement improve, such great uncertainty in the estimation of the Grüneisen parameter has and will continue to diminish.

4. THE MODEL

The currently offered model is not derived from first principles but was developed based on certain functional relationships observed in the UCC model of the cold curve. Though able to effectively match the zero-degree results of the UCC model, the current model is not, in fact, an augmented UCC model. It is, rather, a complete equation of state, consistent with the Grüneisen constraint. By comparison, the UCC model comprises an empirical fit to the zero-degree isotherm, expressed in terms of a nondimensional lattice spacing. It embodies mechanical rather than thermodynamic concepts.

Since p_{ref}, E_{ref}, and ψ are functions of volume alone, the solution to the Grüneisen equation of state [eqn. (8)] amounts to determining ψ and $G(V)$, which satisfy

$$p\psi - E = G(V) \quad . \tag{9}$$

Along the cold curve, where $p_c = -E_c'$, one obtains the differential equation

$$\psi E_c' + E_c = -G(V) \quad . \tag{10}$$

The complementary solution to eqn. (10), corresponding to $G(V) = 0$, is

$$E_c = C\Theta \quad , \tag{11}$$

which may be verified through the use of eqn. (7), where C is an integration constant. Incidentally, it is this complementary solution that constitutes the cold curve in both the Einstein and the Debye equations of state (*i.e.*, no particular solutions were offered). But if E_c involves terms in Θ, it is not unreasonable to expect the same of $G(V)$. A particular solution to the equation

$$\psi y' + y = \Theta \tag{12}$$

is $y(V) = -\Theta \ln\Theta$.

It was the knowledge of these facts that resulted in the inspirational leap of looking at the UCC model (eqn. [1]) and surmising that the a variable was actually a $-K\ln\Theta$ term. Such a speculation was also consistent with eqn. (2), in that the natural logarithm may, over a limited domain, be well approximated by a cube-root relation like eqn. (2). Furthermore, it was speculated that the cubic term of eqn. (1) was a higher-order correction to an incorrect functional form and that it would not be needed if the logarithm were the correct form. Such a speculation could only have originated out of ignorance, of course, when it was realized that the UCC model fits data out to $a = 8$, where the cubic term carries three times the weight of the linear term. As a side note, it was later realized that Vinet *et al.* [2, 3] dropped the cubic term of Rose *et al.* from their formulation as well, since it complicated the simplicity of the form, and was not particularly important in compression. In their case, however, the resulting simplification does adversely affect the quality of their fit to binding energy in the tensile region. As is seen later, such is not the case here, since the use of the logarithmic term does, in fact, compensate for the loss of the cubic term.

Combining these hypotheses yields

$$E_c = E_b\left[1 - (1 + f)\exp(-f)\right] \quad , \tag{13}$$

where

$$f = -K\ln(\Theta/\Theta_0) \quad . \tag{14}$$

Substituting for f gives the cold energy, and the negative of its volume-derivative gives the cold pressure, as

$$E_c = E_b\left[1 - (1 - K\ln(\Theta/\Theta_0))\,(\Theta/\Theta_0)^K\right] \tag{15}$$

and

$$p_c = \frac{E_b K^2}{\psi}\,(\Theta/\Theta_0)^K\,\ln(\Theta/\Theta_0)\quad . \tag{16}$$

The general equation of state that results is obtained, using this cold curve as the reference in eqn (8), to get

$$p\psi - E = E_b\left\{\left[(\Theta/\Theta_0)^K - 1\right] + K(K-1)\,(\Theta/\Theta_0)^K\,\ln(\Theta/\Theta_0)\right\}\quad . \tag{17}$$

It can be seen that eqn (17) becomes greatly idealized if and when the parameter K takes on a value of unity. In order to satisfy the ambient sound speed condition, the constant K must take on a value of

$$K = \frac{C_0}{\Gamma_0\sqrt{E_b}}\quad , \tag{18}$$

which incidentally is $1/(3\Gamma_0)$ times the anharmonicity factor, η, of Rose *et al.* [1] It is shown later that values of K for real metals do, in fact, cluster in the vicinity of unity. As such, the deviation of *K* from unity tells something about the nonidealness of the material. Eqns. (17) and (18) constitute the proposed equation of state, with eqns. (15) and (16) defining the associated cold curve. All that is required at this point is a specification of (Θ/Θ_0) as a function of volume.

5. A USEFUL SIMPLIFICATION

Segletes [10–12] has examined the thermodynamic stability (and instability) associated with various functional forms of the Grüneisen parameter. He proposed a form that could be made not to violate the stability metrics that he examined at the time. He expressed the form, in terms of parameter β, as

$$\Gamma = \frac{\Gamma_0}{1 + \beta\mu} \quad . \tag{19}$$

The family of functions that satisfy this form may be simply expressed another way, in terms of the ψ variable, as

$$\psi'' = 0 \quad . \tag{20}$$

Assuming that this form can adequately characterize metals over some relevant domain of specific volumes, we may then employ it to help us define our needed function (Θ/Θ_0). As was shown when solving eqn. (7) for various Grüneisen forms, the effect of assuming the validity of eqn. (20) provides us with the needed relationship

$$\Theta\,\psi^{(1/\psi')} = \text{constant} \quad , \tag{21}$$

keeping in mind that ψ' and thus the exponent are constants. Thus, we have

$$\Theta/\Theta_0 = (\psi/\psi_0)^{-1/\psi'} \quad , \tag{22}$$

where ψ is linear in V, as dictated by the assumption of eqn. (20). Expressing the proposed cold curve in light of eqn. (22) yields

$$E_c = E_b \left\{ 1 - \left[1 + (K/\psi') \ln(\psi/\psi_0) \right] (\psi/\psi_0)^{(-K/\psi')} \right\} \tag{23}$$

and

$$p_c = - \frac{E_b K^2}{\psi \psi'} (\psi/\psi_0)^{(-K/\psi')} \ln(\psi/\psi_0) \quad . \tag{24}$$

The equation of state becomes

$$p\psi - E = E_b \left\{ \left[(\psi/\psi_0)^{(-K/\psi')} - 1 \right] - (K/\psi')(K-1)(\psi/\psi_0)^{(-K/\psi')} \ln(\psi/\psi_0) \right\} \quad . \tag{25}$$

6. COMPARISON TO THE UCC MODEL

Rose *et al.* [1] were able to express their relationship for the cold energy in terms of parameter a, which, in turn, is expressible in terms of the anharmonicity, η, and the relative volume, V/V_0. The correspondence to the current model cannot be made on a one-to-one basis, as the current model has actually introduced an additional variable. The term ψ/ψ_0, because of the assumed linearity of ψ, may be shown to be

$$\psi/\psi_0 = 1 - (\Gamma_0\psi')\,(1 - V/V_0) \quad . \tag{26}$$

Similarly, the constant multiplier of K/ψ' in the f parameter, eqn. (14), may be re-expressed in terms of η such that f may be expressed as

$$f = \frac{\eta}{3\,(\Gamma_0\psi')}\,\ln\Big(1 - (\Gamma_0\psi')\,(1 - V/V_0)\Big) \quad . \tag{27}$$

Thus, given values for the two parameters η and $\Gamma_0\psi'$, the variables f [eqn. (27)] and a [eqn. (2)] may be implicitly related as a function of volume. The cold energy of the two models, eqns. (1) and (23), may then be compared as well. The $\Gamma_0\psi'$ term determines the effective rate of change of the Grüneisen parameter with volume. For $\Gamma_0\psi' = 1$, the Grüneisen parameter is a constant value of Γ_0, while for $\Gamma_0\psi' = 0$, the Grüneisen function declines with increasing compression at its maximum thermodynamically stable rate, as determined by Segletes [11], according to the function $\Gamma_0/(1 + \mu)$. In all cases of expansion, the Grüneisen parameter asymptotes, at large expansion, to a value of $1/\psi'$. The family of Grüneisen functions determined by eqn. (20) is plotted versus specific volume in Fig. 1, with the parameter $\Gamma_0\psi'$ varying from 0 to unity in increments of 0.2.

In order to preserve the presentation appearance of the UCC model, data are plotted as $(E_c/E_b - 1)$ versus parameter a. In the first series of figures (Figs. 2a–2g), a family of curves, for the current model, is plotted for a given anharmonicity, η, varying $\Gamma_0\psi'$ from 0 to 1, at increments of 0.2. In the second series of figures (Figs. 3a–3e), the parameter $\Gamma_0\psi'$ is specified, and the family of curves, for the current model, is generated by varying the anharmonicity between integral values of 3 and 9, which are seen to cover the full range of values noted by Rose *et al.* (most elements fall in the 4 to 6 range of anharmonicity). In all figures, the UCC model is plotted as a single heavy line. Many observations may be noted about the comparison of the UCC model with the currently proposed model. An enumerated list is given as follows:

1) The general shape of the cold curve for the two models is very similar in form, thereby indicating that, in the current model, the logarithmic function of f seems to compensate for the now-missing a^3 term;

2) From Figs. 2, the curves generated for the current model, corresponding to values for $\Gamma_0\psi'$ in the 0 to 0.4 range, match the UCC model most poorly, in both the tensile ($a > 0$) and compressive ($a < 0$) domains.

3) From Figs. 2b–2d, for anharmonicity ranges of 4 to 6 (87% of elements listed by Rose *et al.* [1] fall in the η=3.5 to 6.5 range), a value of $\Gamma_0\psi'$ in the 0.7 to 0.8 range matches the UCC tensile model very well (see also Figs. 3b, 3c).

4) From Fig. 3c, a value of $\Gamma_0\psi' = 0.8$ matches the UCC model in compression for all values of anharmonicity.

5) From Figs. 2g and 3e, the higher the level of anharmonicity, the greater the value of $\Gamma_0\psi'$ required to match the UCC model results in tension. The best-fit value for $\Gamma_0\psi'$ approaches unity as η increases to a value of 9.

6) From Fig. 2a, for very low values of η equal to 3, it is difficult to pick a single value of $\Gamma_0\psi'$ that matches the UCC results in tension, the required value changing from 0.8 at small expansions to roughly 0.6 at larger expansions.

We may conclude that the simplification afforded by the assumption of eqn. (20) is justifiable for studying the behavior of model over limited domains of compression and expansion, based on the quality of the match to the UCC model. The general model, however, retains the flexibility to permit other functional forms on the Grüneisen parameter and will be explored more fully in the future.

7. COMPARISON TO DATA

The current model, incorporating the simplifying assumption of eqn. (20), has been used to study compression data for various materials. Since the current model is a full equation of state, its predictions may be compared not only for cold-curve compression, but also for other thermodynamic processes (*e.g.*, shock loading, for which ample data exists [18, 19]).

Though it can be done, the comparison intentionally avoids taking the approach of *tuning* all available parameters so as to best correlate the model to both cold and shock data for several reasons. First, it avoids the criticism of so-called "knob turning" to match data. Of course, the current model does have a single parameter, and that is the value of $\Gamma_0\psi'$.

Secondly, it is known that the Grüneisen assumption becomes invalid at large enough pressures, and so an examination of when the current model diverges from the shock data could be used to indicate something about the range of validity of the Grüneisen assumption.

When possible, model parameters were derived directly from the molecular weights as well as the η and ΔE values given by Rose *et al.* [1]. These derived parameters include E_b and C_0. With the addition of the handbook value for the thermal expansion coefficient, α, values for Γ_0 and K are directly obtained. Densities were set to the values from which most or all of the experimental data were obtained.

The only parameter that was subject to fitting was the value of $\Gamma_0\psi'$, which determines the manner in which the Grüneisen function changes with volume (recall Fig. 1). This parameter was fitted not to give the best overall match to both the shock and cold-curve data. Rather, this parameter was fitted to give the best fit to the cold-curve alone. In this way, the model's Hugoniot is merely a prediction of and not an explicit fit to the Hugoniot data. This distinction is important, as better fits to the aggregate cold/Hugoniot data are obtainable by varying values of C_0 and Γ_0 within previously published ranges.

The model data are collected in Table 1. The parameter W is simply the molecular weight, and is used for converting molal quantities to a specific (*i.e.*, per-mass) basis. The second to last column, $\Gamma_0\psi'$, is the only free parameter. Its value reflects the best fit to the cold-curve data. In the last column, ψ', which is also dimensionless, is derived from other column data and is provided for informational purposes only. Graphical results are shown in Figs. 4–12 for a variety of metals. The lower curve is the fitted cold pressure curve. The upper curve (when present) is the predicted Hugoniot curve. Cold data (including theoretical predictions thereof) depicted in the figures came from Mao *et al.* [22], or were digitized from the graphs of Rose *et al.* [1] and Molodets [23]. All shock data, against which the model predictions were compared, were taken from the Livermore compendium [18], the Kohn compilation [19], or Mitchell *et al.* [24]. All cold data are depicted with solid-filled circular symbols. All other (unfilled) symbols represent Hugoniot data.

Table 1. *Model Parameters*

	ρ_0 (kg/m^3)	W (g/mol)	η	E_b [†] (MJ/kg)	C_0 [§] (m/s)	α [‡] (x10^6) (°C^{-1})	Γ_0 [††]	K [‡‡]	$\Gamma_0 \psi'$	ψ'
Ag	10490	107.868	5.94	2.647	3221	19.0	2.55	0.78	0.84	0.33
Al	2700	26.982	4.71	11.939	5424	25.0	2.39	0.66	0.81	0.34
Cu	8930	63.546	5.20	5.312	3995	16.6	2.02	0.86	0.78	0.39
Li	537	6.941	3.10	22.928	4948	-	-	-	0.81	-
Mo	10200	95.94	5.85	6.846	5102	5.0	1.50	1.30	0.43	0.29
Pd	12020	106.4	6.41	3.568	4036	11.7	2.44	0.88	0.61	0.25
Pt	21430	195.09	6.47	2.893	3515	9.0	2.61	0.79	0.78	0.30
Rb	1530	85.47	4.18	0.968	1371	-	-	-	0.88	-
304 St. [¶]	7896	56.05	5.24	7.342	4733	12.0	1.81	0.97	0.76	0.42

[†] E_b calculated from ΔE values of Rose *et al.* [1] using $E_b(\text{MJ/kg}) = 96.45\,\Delta E(\text{eV})/W$.

[§] C_0 calculated using eqn. (3) and values of η given by Rose *et al.* [1], rather than handbook values. Platinum is the exception, where the value of 3668 m/s yielded by eqn. (3) produces a fit to the cold curve that is not as good as when using the handbook value of 3515 m/s.

[‡] Values for thermal expansion coefficient taken from Weast [20], except for palladium, which was taken from Layne [21], and lithium and rubidium, which could not be located in the literature.

[††] For consistency, initial Grüneisen values computed using $\Gamma_0 = \alpha C_0^2 W/R$, where R is the universal constant, equal to 8314 g m^2/s^2 mol K. The values for Γ_0 are generally compatible with published values found elsewhere. Values of α for lithium and rubidium could not be located (nor could estimates of Γ_0).

[‡‡] K calculated using eqn. (18).

[¶] Properties for 304 stainless steel calculated, per Rose *et al.* [1], using 70% Fe, 19% Cr, 11%Ni.

The fits to the cold-compression data are extremely good, with the model curve literally overlaying the data in nearly every case. For most cases, the Hugoniot predictions are very good to a point, though model results in some cases, notably silver and platinum, diverge from data at higher pressures. Lithium and rubidium are special cases, in that it does not appear possible that the cold [1] and Hugoniot [18] data were even generated from the same density base material. In particular, Hugoniot data, which should universally lie at higher pressures than the cold curve, are not seen to exceed the cold-curve data until a relative compression of approximately 2. For this reason, and the fact that thermal expansion data and Grüneisen estimates could not be obtained for the materials, no attempt was even made to model the lithium and rubidium Hugoniots. In the case of aluminum, the poor fit at even moderate pressures is perhaps attributable to a phase change since, as shown in Figure 5 with the dotted curve, when using an initial Grüneisen value of 1.2 (half of the value shown in Table 1), an excellent fit to the aluminum Hugoniot prevails to 9 Mbar. Such a notion is buttressed by the work of McKenna and Pastine [16], whose empirical fit to the aluminum Grüneisen function decreases with compression at such a prodigious rate (in the V/V_0 range from 1.0 to 0.85) that it violates the stability metrics of Segletes [10, 11], established for thermodynamic transitions of single-phase Grüneisen materials. Thus, if McKenna's fit is accurate, then either the aluminum Grüneisen function varies with temperature (violating the Grüneisen assumption) or else the aluminum Hugoniot experiences a phase change in V/V_0 range of 0.85 to 1.0.

Table 2 gives a very approximate estimation of the goodness of the match of the current model to the Hugoniot data, in terms of pressure, and includes, in the last column, the Debye temperature for each material. In all cases except aluminum, good matches to the Hugoniot are obtained out to pressures of approximately 1 Mbar or greater. Furthermore, the two materials, silver and platinum, that were observed to experience only fair correlations at higher pressures, happen to be the materials with notably low Debye temperatures. It is

Table 2. *Limiting Pressure Ranges (Mbar) to which Model Prediction Qualitatively Correlates with Hugoniot Data*

	Quality of Model Correlation with Hugoniot Data			Θ_D[†] (K)
	Excellent	Good	Fair	
Ag	0.5	0.9	1.6+	220
Al	—	0.5	0.7	380
Cu	1.2	4.2+	—	310
Mo	0.6	1.7+	—	375
Pd	0.5	2.2+	—	—
Pt	2.0	2.0	2.7+	225
304 St. Steel	1.9+	—	—	433[‡]

[†] Data extracted from Andrews [6].

[‡] Value reflects that of pure iron.

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surmised that perhaps the breakdown of the Grüneisen assumption, eqn. (4), at high pressures (actually temperature) is responsible for the divergence of the model from the data. In the case of aluminum, a possible cause of the poor Hugoniot prediction has already been discussed. In line with specific heat theory and the notion of a characteristic temperature, it would seem to make proper thermodynamic sense that the breakdown of the Grüneisen assumption with temperature might also scale with the characteristic temperature [*i.e.*, the breakdown occurs as a function of (Θ/T)]. With the exception of aluminum, the results of Table 2 would seem to support this reasoning.

8. CONSTRAINTS ON THE FREE PARAMETER, $\Gamma_0 \psi'$

The free parameter in the current model, $\Gamma_0 \psi'$, obviously has physical meaning and relates to the rate of Grüneisen parameter change with volume. Ideally, it could be quantified from other physical constants. Since the characteristic temperature in the current model is always normalized by its ambient value though, one is limited in trying to extract additional information directly from this form. The assumed Grüneisen form, eqn. (20), dictates that $\Gamma_0 \psi' = 1$ be an upper limit on the model parameter. For a lower limit, stability analyses conducted by Segletes [11] showed, on the basis of high pressure isobaric transitions, that the maximum rate of descent of the Grüneisen function with compression is limited such that $\psi' \geq 0$. At cold pressures, however, a different isobaric criterion arises, in which it was shown that a stable compressive cold curve must obey the relation

$$-p_c'/p_c \geq 1/\psi \quad . \tag{28}$$

Applying this criterion to the current model gives a lower bound on the model parameter as

$$\psi' \geq \frac{1 - K}{1 - \dfrac{1}{\ln(\psi/\psi_0)}} \quad . \tag{29}$$

It should be immediately noted that the value of the parameter K, which equals $\eta/3\Gamma_0$, drives the lower bound on ψ'. In particular, for materials in which K is greater than unity, the high-pressure $\psi' \geq 0$ criterion is the most restrictive constraint. For K less than unity, the low-pressure constraint, given by eqn. (29), governs the problem. For K equal to unity, both criteria coincide and the equation of state, eqn. (17), simplifies immensely, with the right-hand term identically zero and all exponents equal to unity.

Considering eqn. (29), for the worst-case compression scenerio, one may take the argument of the logarithm as $(1 - \Gamma_0 \psi')$. The results of this criterion give lower allowable limits on the model parameter, $\Gamma_0 \psi'$, below the fitted value for each of the materials studied, with minimum levels generally below the 0.4 level. This result reinforces our comparison with the UCC model, in which low values (0 to 0.4) of the model parameter did not fit the UCC model in tension well. The span from 0.4 to 1, though, still leaves a large, permissable range on the value of the parameter $\Gamma_0 \psi'$. Further analysis may serve to better define this parameter, either in terms of more restrictive limits or in terms of fundamental quantity groupings.

9. CONCLUSIONS

An equation of state for metals has been proposed. The form of the equation provides a very close match to the cold-temperature expansion models, cold-compression data, and shock-compression data. The model was inspired by the Universal Cold Curve (UCC) model of Rose *et al.* [1], when it was realized that algebraic terms in the UCC model bore a functional resemblence to thermodynamic terms that could be the solution to the Grüneisen cold curve. In the model, the cold energy is completely expressible with terms involving the material's characteristic temperature.

A simplified form of the current model has one free parameter per given material, which physically relates to the rate at which the Grüneisen parameter changes with volume.

The fitting parameter for the model is not totally free, but is constrained within certain bounds, based on considerations of thermodynamic stability. With this one parameter, excellent fits to cold curves are produced to all available low-temperature compression data, including platinum cold compressions to 6 Mbar. Once the fitting parameter is set for the cold curve, other thermodynamic paths may be predicted without the need of additional fitting parameters. The resulting predictions of Hugoniot behavior match shock-compression data well, typically to the megabar range, before diverging. The divergence of the current model with very high pressure shock data might be attributable to the breakdown of the Grüneisen assumption at high pressures.

Additional avenues remain to be explored with the current model, which include enhancing the formulation to properly account for the very high pressure regions, examining more fully the properties of the equation of state in tension, and examining the general form of the equation of state, eqn. (15). Ultimately, one seeks to understand the underlying physics of lattice compression and vibration that would lead to an equation of state of the form proposed. As it is, the current model hopefully serves to put the often-used Grüneisen equation of state on a firmer foundation.

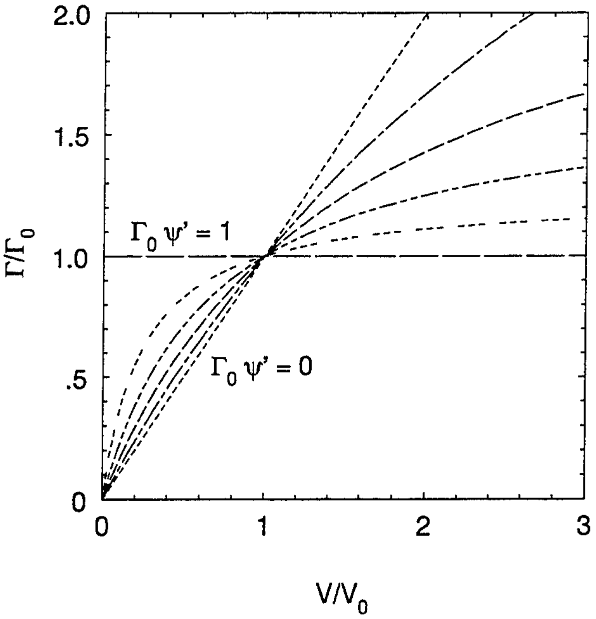
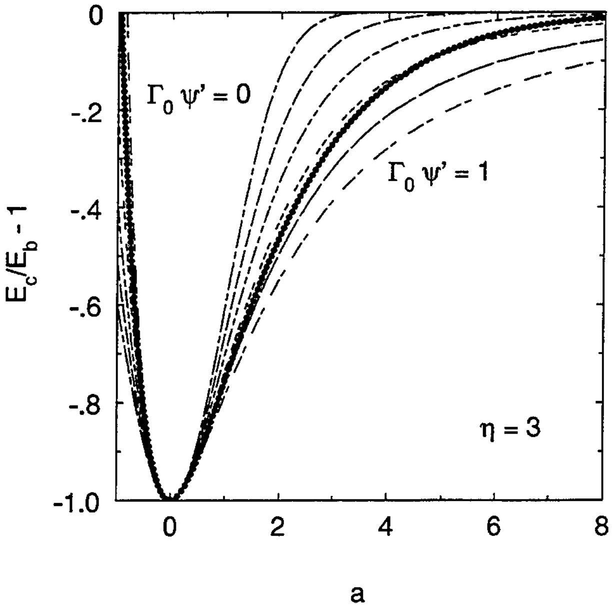
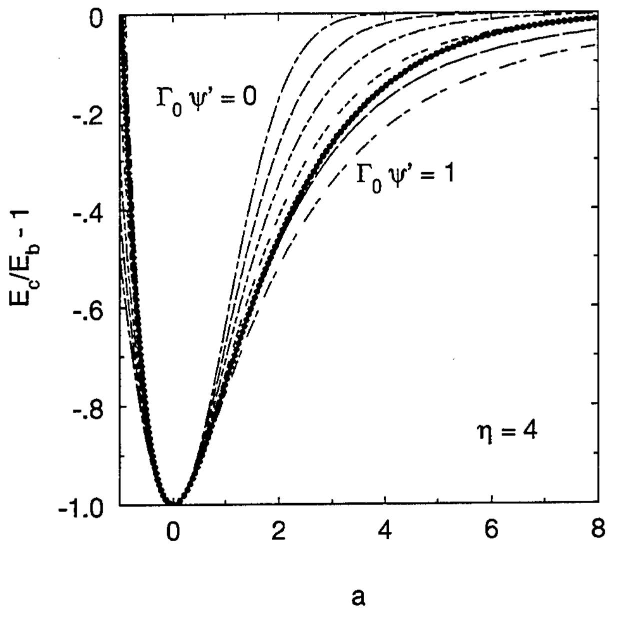


Figure 1. *Relative Grüneisen parameter versus relative volume as a function of parameter* $\Gamma_0\psi'$*, taken in increments of 0.2, for the assumed form* $\psi'' = 0$.

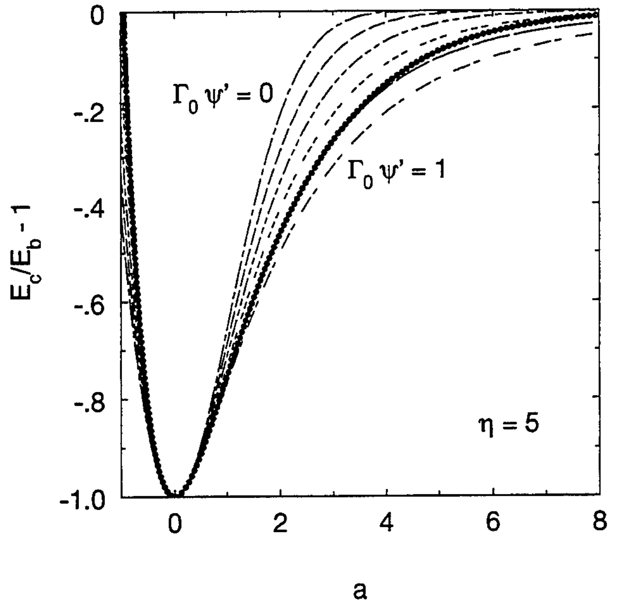


(a)

Figure 2. *Comparison of binding energy of current model to that of Rose et al. [1], for given anharmonicities. Rose curve depicted with dark line. Current model shown in dashed lines for incremental values of* $\Gamma_0\psi'$*, ranging from 0 to 1 in increments of 0.2. (a)* $\eta = 3$. *(b)* $\eta = 4$. *(c)* $\eta = 5$. *(d)* $\eta = 6$. *(e)* $\eta = 7$. *(f)* $\eta = 8$. *(g)* $\eta = 9$.

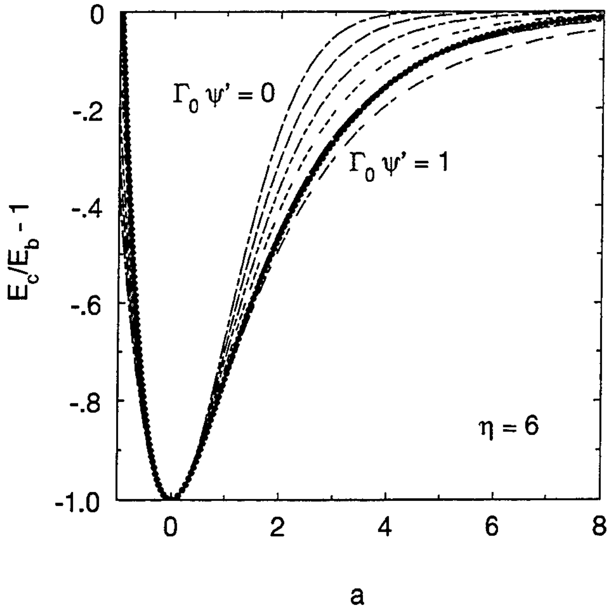


(b)



(c)

Figure 2 (continued). *Comparison of binding energy of current model to that of Rose et al. [1], for given anharmonicities. Rose curve depicted with dark line. Current model shown in dashed lines for incremental values of $\Gamma_0\psi'$, ranging from 0 to 1 in increments of 0.2. (a) η = 3. (b) η = 4. (c) η = 5. (d) η = 6. (e) η = 7. (f) η = 8. (g) η = 9.*



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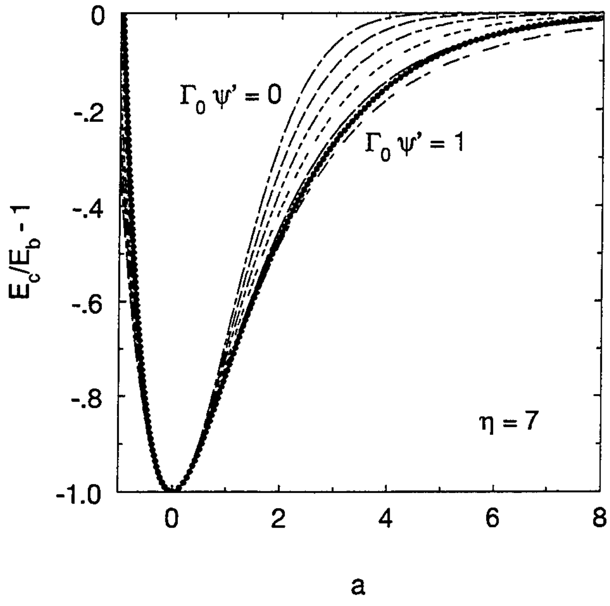
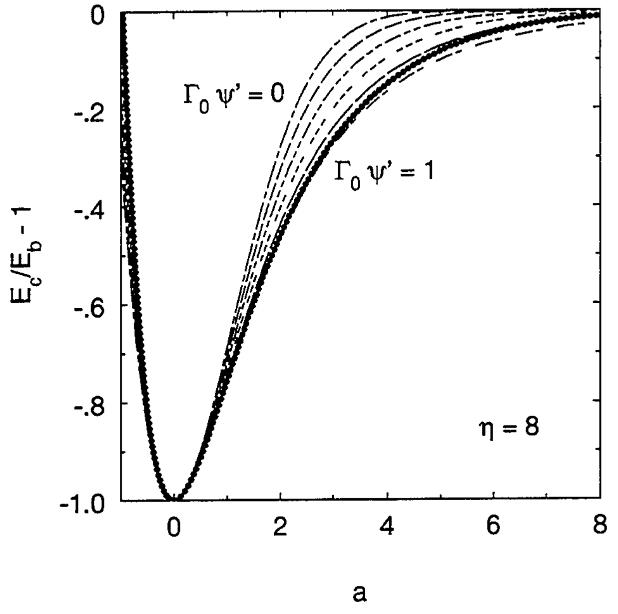


(e)

Figure 2 (continued). *Comparison of binding energy of current model to that of Rose et al. [1], for given anharmonicities. Rose curve depicted with dark line. Current model shown in dashed lines for incremental values of* $\Gamma_0 \psi'$, *ranging from 0 to 1 in increments of 0.2. (a)* $\eta = 3$. *(b)* $\eta = 4$. *(c)* $\eta = 5$. *(d)* $\eta = 6$. *(e)* $\eta = 7$. *(f)* $\eta = 8$. *(g)* $\eta = 9$.



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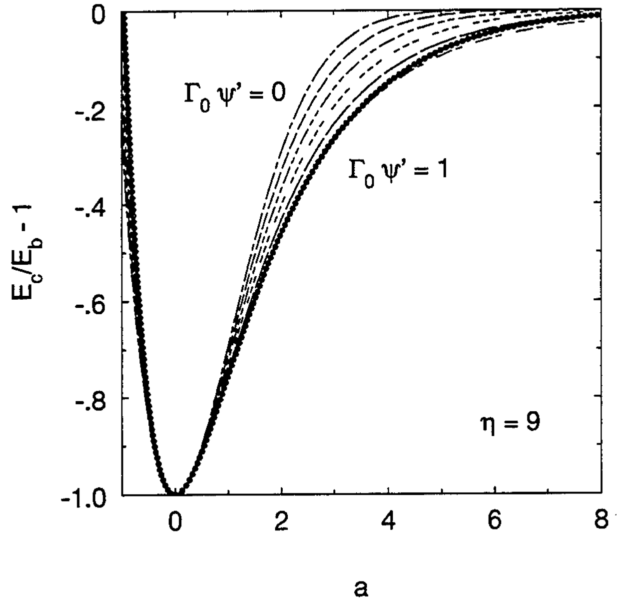
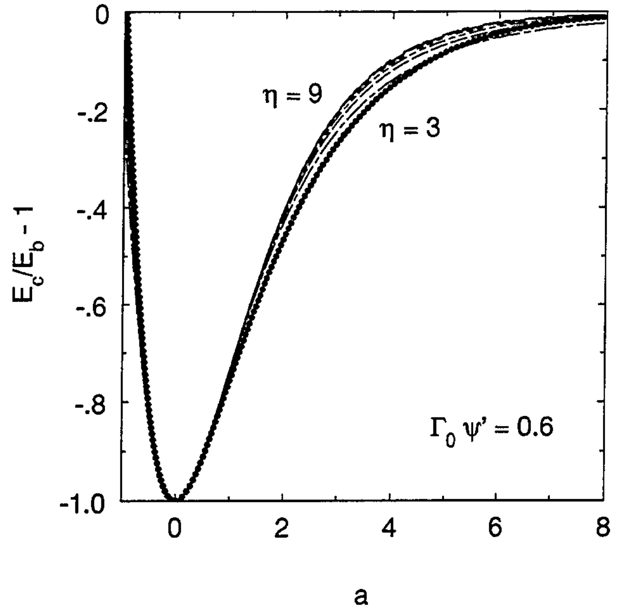


(g)

Figure 2 (continued). *Comparison of binding energy of current model to that of Rose et al. [1], for given anharmonicities. Rose curve depicted with dark line. Current model shown in dashed lines for incremental values of* $\Gamma_0\psi'$*, ranging from 0 to 1 in increments of 0.2. (a)* $\eta = 3$*. (b)* $\eta = 4$*. (c)* $\eta = 5$*. (d)* $\eta = 6$*. (e)* $\eta = 7$*. (f)* $\eta = 8$*. (g)* $\eta = 9$*.*



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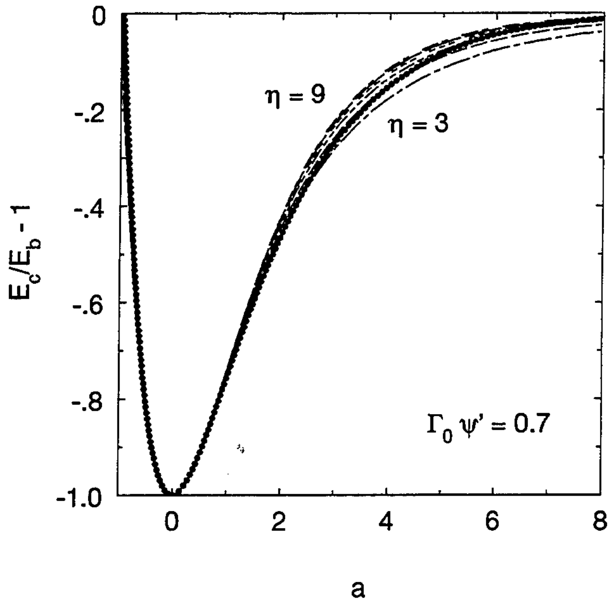
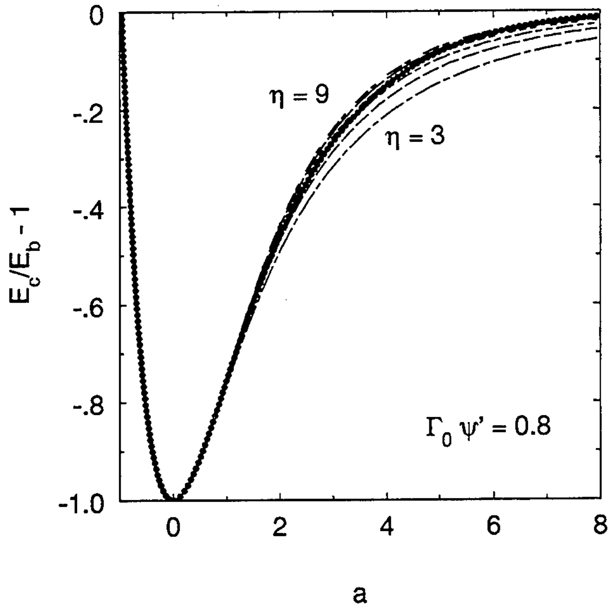


(b)

Figure 3. *Comparison of binding energy of the current model to that of Rose et al. [1], for given values of $\Gamma_0\psi'$ parameter. Rose curve depicted with dark line. Current model shown in dashed lines for integral values of η, ranging from 3 to 9.*
(a) $\Gamma_0\psi'$ = 0.6. (b) $\Gamma_0\psi'$ = 0.7. (c) $\Gamma_0\psi'$ = 0.8. (d) $\Gamma_0\psi'$ = 0.9. (e) $\Gamma_0\psi'$ = 1.0.



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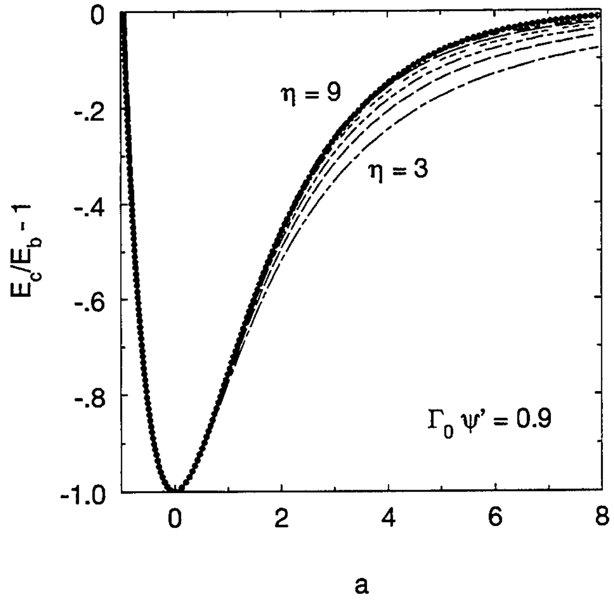
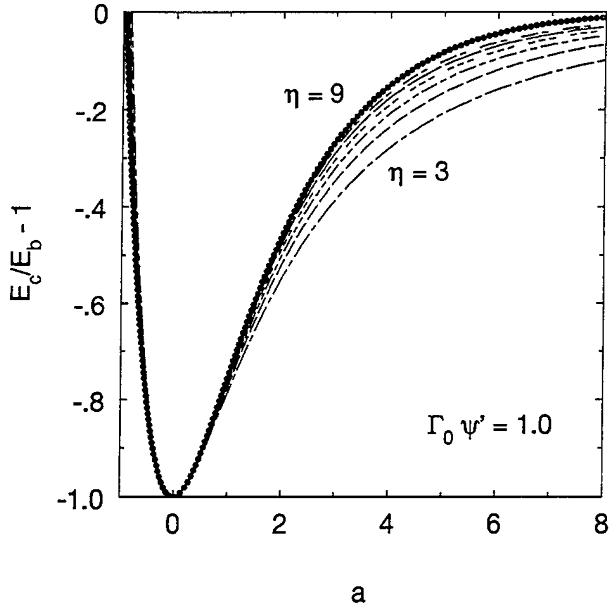


(d)

Figure 3 (continued). *Comparison of binding energy of the current model to that of Rose et al. [1], for given values of* $\Gamma_0\psi'$ *parameter. Rose curve depicted with dark line. Current model shown in dashed lines for integral values of* η, *ranging from 3 to 9.* *(a)* $\Gamma_0\psi' = 0.6$. *(b)* $\Gamma_0\psi' = 0.7$. *(c)* $\Gamma_0\psi' = 0.8$. *(d)* $\Gamma_0\psi' = 0.9$. *(e)* $\Gamma_0\psi' = 1.0$.



(e)

Figure 3 (continued). *Comparison of binding energy of the current model to that of Rose et al. [1], for given values of $\Gamma_0 \psi'$ parameter. Rose curve depicted with dark line. Current model shown in dashed lines for integral values of η, ranging from 3 to 9. (a) $\Gamma_0 \psi'$ = 0.6. (b) $\Gamma_0 \psi'$ = 0.7. (c) $\Gamma_0 \psi'$ = 0.8. (d) $\Gamma_0 \psi'$ = 0.9. (e) $\Gamma_0 \psi'$ = 1.0.*

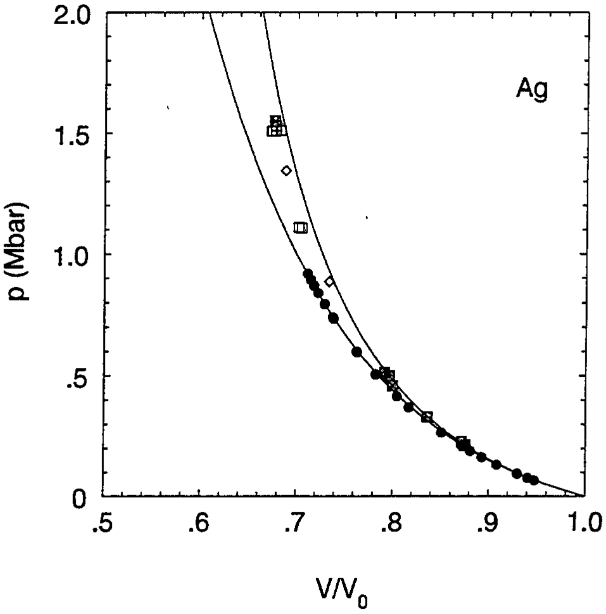


Figure 4. *Cold-curve fit with Hugoniot prediction for silver, to 2 Mbar. Cold data [22] (filled symbols) and Hugoniot data [18, 19] (unfilled symbols) included.*

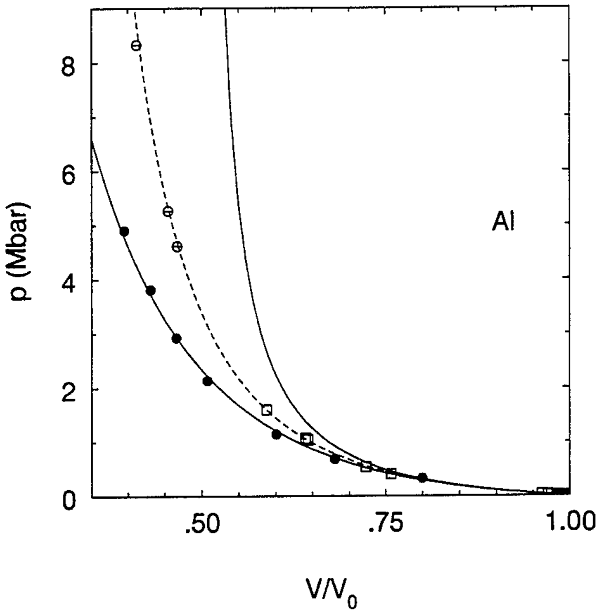


Figure 5. *Cold-curve fit with Hugoniot prediction for aluminum, to 9 Mbar. Cold data [23] (filled symbols) and Hugoniot data [19, 24] (unfilled symbols) included. Dashed curve is Hugoniot prediction, assuming* $\Gamma_0 = 1.2$.

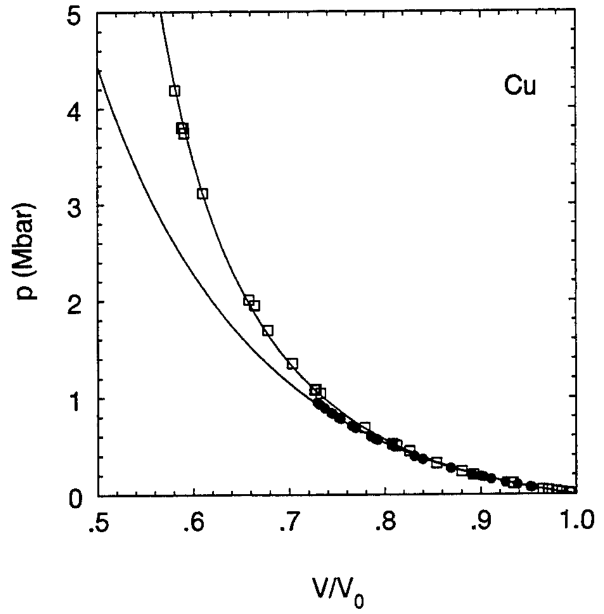


Figure 6. *Cold-curve fit with Hugoniot prediction for copper, to 5 Mbar. Cold data [22] (filled symbols) and Hugoniot data [19] (unfilled symbols) included.*

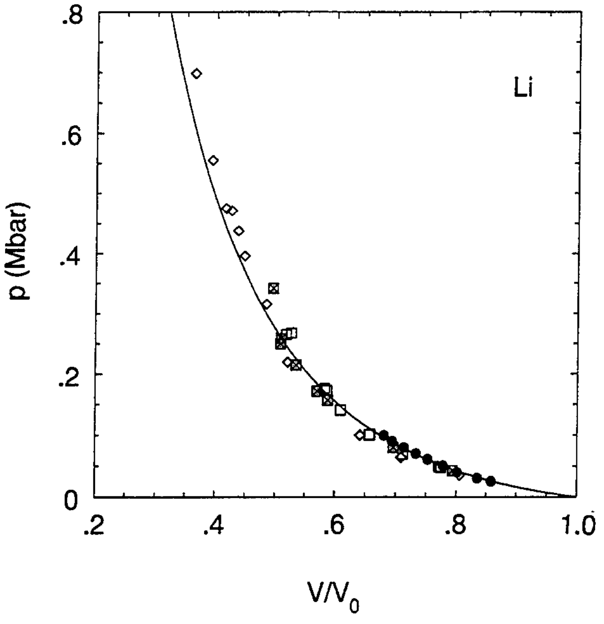


Figure 7. *Cold-curve fit for lithium, to 0.8 Mbar. Cold data [1] (filled symbols) and Hugoniot data [18] (unfilled symbols) included.*

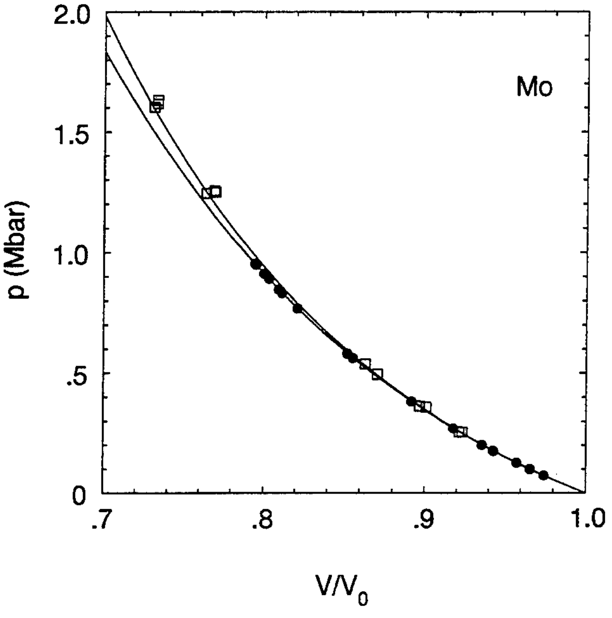


Figure 8. *Cold-curve fit with Hugoniot prediction for molybdenum, to 2 Mbar. Cold data [22] (filled symbols) and Hugoniot data [19] (unfilled symbols) included.*

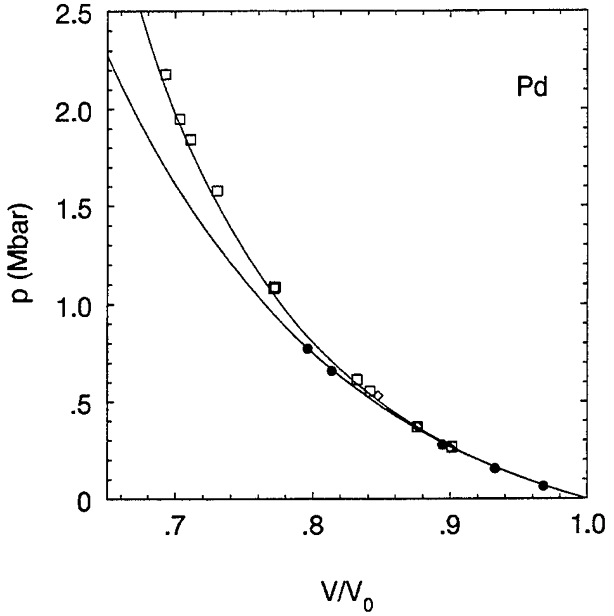


Figure 9. *Cold-curve fit with Hugoniot prediction for palladium, to 2.5 Mbar. Cold data [22] (filled symbols) and Hugoniot data [18] (unfilled symbols) included.*

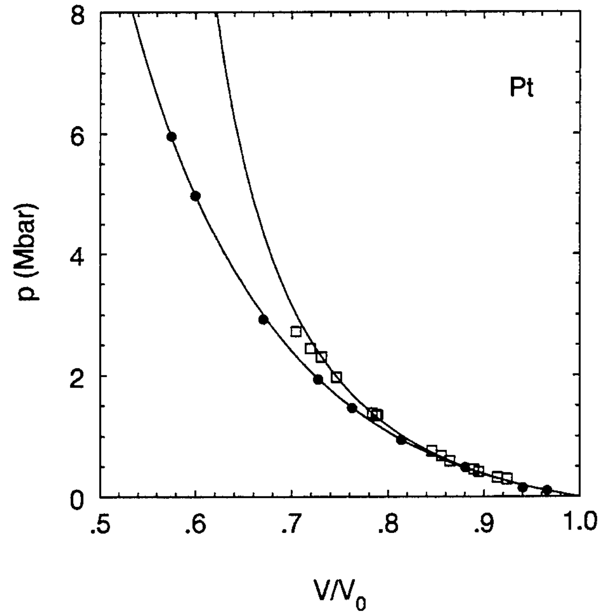


Figure 10. *Cold-curve fit with Hugoniot prediction for platinum, to 8 Mbar. Cold data [1] (filled symbols) and Hugoniot data [19] (unfilled symbols) included.*

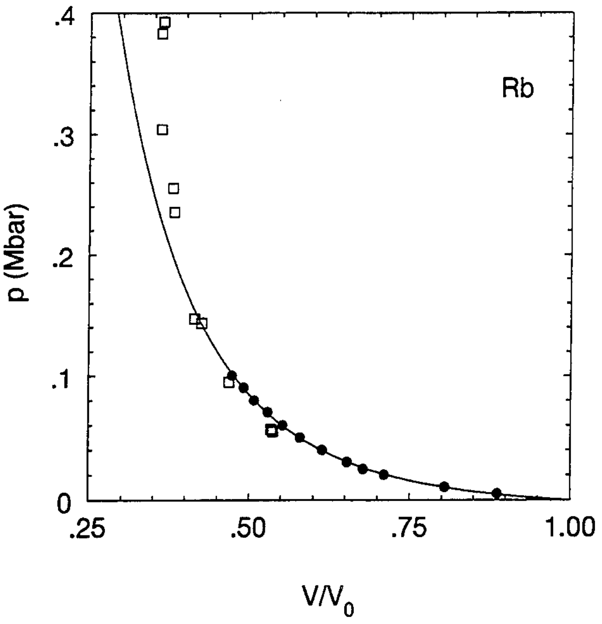


Figure 11. *Cold-curve fit for rubidium, to 0.4 Mbar. Cold data [1] (filled symbols) and Hugoniot data [18] (unfilled symbols) included.*

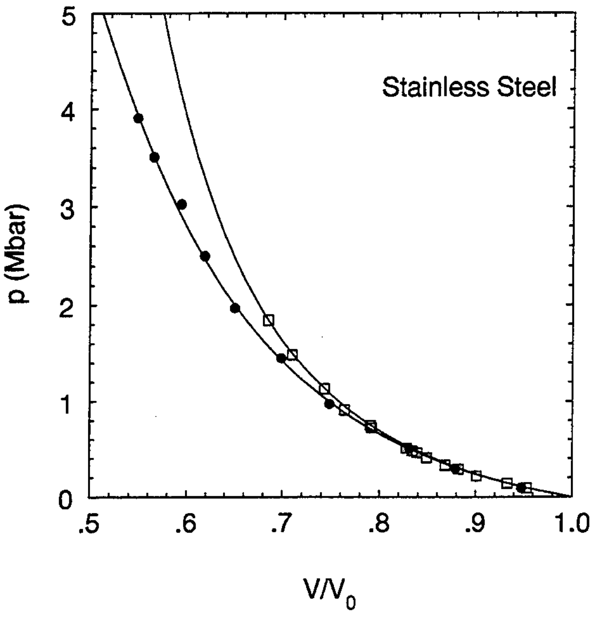


Figure 12. *Cold-curve fit with Hugoniot prediction for 304 stainless steel (70% Fe, 19% Cr, 11% Ni), to 5 Mbar. Cold data [1] (filled symbols) and Hugoniot data [19] (unfilled symbols) included.*

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13. ABSTRACT *(Maximum 200 words)*

A new analytical equation of state is offered for metals. The form is expressible completely in terms of the maximum lattice vibration frequency, macroscopically expressible as the characteristic temperature, Θ, and its derivatives. For one particularly useful special case, the results have been found to match both cold-curve and shock-Hugoniot data for a variety of metals. The equation is of the Grüneisen variety, in which the Grüneisen parameter is specified as a function of volume only. The equation begins to lose accuracy in the pressure range of one to several megabars, perhaps because of the breakdown of the Grüneisen assumption, whereupon the Grüneisen parameter becomes energy dependent.

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