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On Theories of the Grüneisen Parameter

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and William P. Walters

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On Theories of the Grüneisen Parameter

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Abstract

A recently proposed equation of state is further examined for compatibility with prior theory. It is found, by adopting a more realistic functional relationship for one of the thermodynamic state variables and eliminating higher-order terms from the model, that three historical theories for determination of the Grüneisen function fall directly out of the recently proposed model. One is led to conclude that the newer model captures the essence of all of the older theories, each of which being applicable under an appropriate circumstance.

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1. BACKGROUND

The estimation and measurement of the Grüneisen function for metals is an important but difficult task. Simple formulas exist for its estimation, in terms of specific heat and compressibility, or alternately in terms of derivatives of the cold (*i.e.*, zero degree) compression curve, or still again in terms of derivatives of the lattice frequency. Depending on the validity of various assumptions about the material, some of these definitions may be redundant. However, when applied to actual substances, these methods can produce estimates that vary by significant amounts (100%, in some cases [1]), even at ambient conditions.

Some variation is due to the fact that the Grüneisen assumption, which has the Grüneisen function independent of temperature, is only an approximation. It is well known [2,3], for example, that at low temperatures (at fractions of the Debye temperature) the Grüneisen parameter is indeed temperature dependent. Grodzka [1] presents data showing a significant thermal variation in the Grüneisen parameter at higher temperatures as well, for some materials. Other variations in the estimate arise out of the differences in modeling of the propagation velocities associated with lattice vibration. Three widely referenced models of this variety, which provide not only an ambient value but also a volume-dependent estimate of the Grüneisen function, are those of Slater [4], Dugdale and MacDonald [5], and the free-volume theory [6]. Pastine [7] and Vashchenko and Zubarev [6] have attempted to generalize results by showing that the variation of Poisson's ratio with volume can account for some of the differences in the various models of the Grüneisen parameter. Unfortunately, experimental knowledge of how Poisson's ratio varies with compression and/or temperature is as elusive as knowledge of the Grüneisen function itself.

Historically, the estimation of the Grüneisen function was further complicated by the paucity of cold-compression data. Available high-pressure data were almost exclusively that of shock transition, and so the cold-compression curves, needed by the various models in order to estimate the Grüneisen function, were necessarily back-extrapolated from shock data using a Grüneisen function that was *a priori* unknown (*e.g.*, see Walsh *et al.* [8]).

To some extent, this uncertainty in the functional behavior of the Grüneisen function has persisted to the present. For example, Segletes [9] examined the Grüneisen formulations present in five of the widely used hydrocodes, which were developed by the U.S. Department of Energy (DOE) national labs and the U.S. Department of Defense (DOD), as well as industry. In examining these codes, which are utilized to model the high-compression deformations associated with high-velocity impact and/or explosive loading, it was noted that a common Grüneisen formulation was not employed. Not only were the Grüneisen formulations different from code to code, but Segletes was able to show, in some cases, that thermodynamic instabilities would result because of an incompatible selection of Grüneisen and Hugoniot functions.

In a significant work, which, on the surface, might not appear to shed light on the problem of estimating the Grüneisen function, Rose *et al.* [10] proposed (in 1984) a universal cold curve, which was able to provide a generic function for the lattice binding energy, which could be appropriately scaled to match theoretical and experimental data for many metals. Since the work of Rose and his colleagues, others have continued in the area as well, notably the same team led by Vinet [11,12] and, more recently, a group led by Baonza [13,14]. These later efforts succeeded in introducing temperature effects and thus produced a Grüneisen-style equation of state. However, in all of these more recent efforts cited, the function governing the reference-compression curve was mutually exclusive of the function used to model the thermal effects. In the works of Vinet *et al.*, the variation of the bulk modulus was fitted for a number of materials, which effectively parlayed into a Grüneisen function. In the case of Baonza *et al.*, a particular, commonly employed, functional form of the Grüneisen function was used—namely, Γ/V equals a fitted constant. While all of these works are worthy efforts in their own right, their primary contribution lies in an area other than the theory of the Grüneisen function.

Segletes [15,16] recently proposed an equation of state for metals, which captures, in the absence of phase change, the behavior of metals into the megabar range. The model was likewise inspired by the universal cold-curve work of Rose *et al.* [10]. However, unlike the efforts of Vinet *et al.* [11,12] and Baonza *et al.* [13,14], in which the function governing the cold compression was mutually exclusive of the function used to model the thermal effects, the recent work of Segletes, by expressing the binding-energy function in terms of lattice frequency, as opposed to lattice compression, unified the functions governing cold-compression and thermal effects. In essence, a specification of the cold-compression behavior in Segletes' model automatically yields the Grüneisen function, and conversely, a specification of the thermal effects, by way of the Grüneisen function, automatically yields a cold-compression curve.

Segletes' equation is of the Grüneisen variety and is given by

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

where p and E are the pressure and specific internal energy, respectively, and the constant E_b is the specific binding energy of the lattice. The terms Θ_0 and Θ are the ambient and current values of the characteristic temperature of the lattice, which, according to the tenets of Grüneisen theory, is assumed to be a function of volume only and is proportional to the maximum lattice vibrational frequency, ν. The concepts of characteristic temperature and lattice frequency derive from the early work on specific heats at low temperatures, by Einstein, Debye, Born and von Karman, and others. In the current context, the characteristic temperature serves as a macroscopic parameter that permits the vibrational frequency of the lattice (a microscopic quantity) to be inferred—namely, the theory of specific heats has these two parameters directly proportional. Returning to eqn. (1), the parameter ψ is the thermodynamic state variable equal to the ratio of specific volume to the Grüneisen parameter, V/Γ, introduced by Segletes [17,18] for convenience in manipulating the thermodynamic equations. The constant K is given by

$$K = \frac{C_0}{\Gamma_0 \sqrt{E_b}} \quad , \tag{2}$$

which incidentally is $1/(3\Gamma_0)$ times the anharmonicity of the lattice. Values for K have been observed to cluster in the 2/3 to 4/3 range for metals, which is compatible with the model's behavior becoming greatly idealized (*i.e.*, linear in Θ/Θ_0) when the constant K takes on a value of unity. C_0 is the bulk sound speed at absolute zero temperature and pressure, and Γ_0 is the ambient value of the Grüneisen parameter.

In this model, the cold curve (*i.e.*, zero-degree isotherm) is given by

$$E_c = E_b\left\{1 - \left[1 - K\ln(\Theta/\Theta_0)\right](\Theta/\Theta_0)^K\right\} \tag{3}$$

and

$$p_c = \frac{E_b K^2}{\psi}(\Theta/\Theta_0)^K \ln(\Theta/\Theta_0) \ , \tag{4}$$

where p_c and E_c are the cold pressure and specific cold energy, respectively.

In macroscopic terms, the Grüneisen function, Γ, is defined by $\Gamma = V/\psi = V(\partial p/\partial E)_V$. From microscopic arguments, however, the characteristic temperature, Θ (alternately, the maximum lattice vibrational frequency, ν), is related to the ψ variable through the relationship that defines the Grüneisen parameter:

$$\frac{\nu'}{\nu} = \frac{\Theta'}{\Theta} = -\frac{1}{\psi} \ , \tag{5}$$

where primes (here and throughout the paper) denote ordinary differentiation with respect to specific volume. In the general case, the differentiation in eqn. (5) would be partial at constant temperature. However, for Grüneisen materials, in which $\Gamma = \Gamma(V)$ only, the derivative in eqn. (5) becomes ordinary.

It is interesting to note the distinction between Segletes' equations and those of an ideal, harmonic oscillator, which is often used to approximate a crystal lattice for purposes of theoretical derivation. In the case of Planck's harmonic oscillator at zero temperature [19], $dE_c/d\nu$ is constant. No lattice is truly harmonic, of course, or else its thermal expansion coefficient and Grüneisen function would both be identically zero. From eqns. (3) and (5), it can be seen, for the idealized case where K is unity, that the current model would have $dE_c/d\nu \propto \ln(\nu/\nu_0)$. A theoretical explanation for this distinction is not yet postulated.

Once the ψ function is defined, integration of eqn. (5) gives the characteristic temperature, which is the primary equation-of-state variable in eqn. (1). In the original papers, Segletes [15,16] incorporated a linear relationship for $\psi(V)$, based on results of his prior work on thermodynamic stability [9,17,18], given in general form by

$$\psi/\psi_0 = 1 - (\Gamma_0\psi')(1 - V/V_0) \quad . \tag{6}$$

The ambient values of specific volume and Grüneisen parameter determine ψ_0, while the constant $(\Gamma_0\psi')$ was the sole material parameter that was actually fitted to the cold curve of each metal. The fitted values of $(\Gamma_0\psi')$, for nine metals, ranged from 0.43 to 0.88, with a vast majority clustered in the vicinity of 0.8. In terms of compression, μ, equal to $(V_0-V)/V$, the linear ψ translates to a Grüneisen function of the form $\Gamma=\Gamma_0/(1+\beta\mu)$, with β as the fitting parameter.

This assumed linear relationship produced an excellent match to empirical data and first-principles computations over a very wide range of specific volumes. However, for extremely large compressions, this linear relationship can no longer be expected to hold, as it is later shown that the ψ variable must vanish in the high-compression limit ($V \to 0$), whereas the linear form of eqn. (6) will not do so for values of $(\Gamma_0\psi')$ other than unity.

In this report, a power law for the $\psi(V)$ parameter, to replace the linear relationship originally employed, is adopted. Though this choice of function still suffers deficiencies at very large compressions, a number of positive outcomes result in the context of Segletes' model [15,16] with this change: 1) the $\psi(V)$ parameter vanishes with diminishing specific volume, as is required; 2) binding-energy expansion data for actual metals are better fit with the power law, as compared to the linear assumption; and most importantly, 3) Segletes' equation of state is shown, by neglecting higher-order terms, to reduce to the model of Slater [4], or Dugdale and MacDonald [5], or to the free-volume theory [6], depending on the value of the exponent used in the power law for $\psi(V)$.

2. THE HIGH-COMPRESSION LIMIT FOR $\psi(V)$

With the linear relation for $\psi(V)$, given by eqn. (6), the high-compression limit for ψ approaches a value of $\psi_0\,(1 - \Gamma_0\psi')$, which would typically lie in the $0.2 \cdot \psi_0$ range for metals. To show what high-pressure limit is appropriate for ψ, in the context of the new model, theoretical values for cold pressure and energy using Thomas-Fermi-Dirac (TFD) theory for several metals were inserted into the new model in order to back out values for ψ. In this manner, the high-pressure limiting functional behavior for ψ, which would be required to match TFD theory, was obtained. Values for the TFD data were obtained using the analytical fitting equations described by Kerley [20].

It is not asserted here that the new equation of state can or should capture the high-compression atomic interactions modeled with TFD zero-temperature theory. Rather, it merely makes sense, from an application point of view, that the current model should blend smoothly into TFD theory, so that a computational implementation can smoothly transition between applicable theories.

Results of the TFD fits for ψ are shown in Figure 1, for several metals. Superimposed on the graph are lines of constant $\Gamma_0\psi'$, which characterized the original linear form employed by Segletes [15,16]. A finite limiting value of ψ implies that Γ approaches zero in the high-

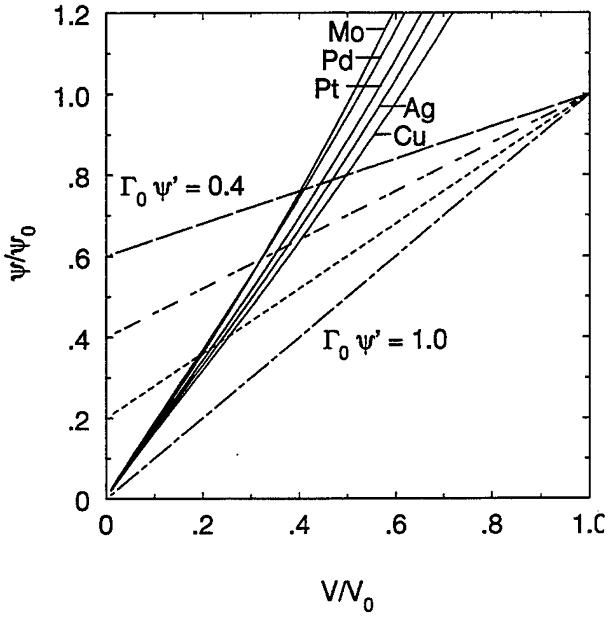

Figure 1. *Limiting ψ values for several metals (solid lines), obtained by inserting TFD cold-curve values into equation of state of Segletes. Superimposed (dashed lines) are limiting ψ values associated with the linear ψ assumption.*

compression limit, which is known not to be the case. The TFD results indicate that ψ must drop to zero with decreasing specific volume, which indicates that the limiting value of Γ is nonzero, but instead related to the limiting slope. Regardless of material, the TFD results are roughly linear, having a slope of approximately 2 in the very-low-volume limit, with the slope diminishing slightly with increasing volume. A finite limiting slope of value m in Figure 1 implies a limiting value of the Grüneisen parameter of Γ_0/m. The fact that (in the case of $m \approx 2$) $\Gamma_0/2$ doesn't equal 1/2 or 2/3, as might be expected, merely indicates that the current model fails to capture the essence of TFD theory, which is no great surprise since the model's equations are not those of TFD theory. In any event, the linear treatment of ψ, originally employed, does not blend smoothly into the realm where TFD theory governs. Another functional choice for ψ might be more appropriate.

3. A POWER LAW FOR $\psi(V)$

As an alternative to the linear relationship for $\psi(V)$, given by eqn. (6), a power law of the form

$$\psi/\psi_0 = (V/V_0)^x \tag{7}$$

is investigated. The power-law form for $\psi(V)$ retains the mathematical simplicity necessary to manipulate the governing equations conveniently, and despite a few deficiencies that will be pointed out, it seems to be a step in the right direction. In terms of compression, μ, the power-law ψ translates to a Grüneisen function of the form $\Gamma=\Gamma_0/(1+\mu)^{1-x}$.

Figure 2 shows the same TFD results as presented previously, except that several contours of a power-law $\psi(V)$ are superimposed on the graph, rather than the linear contours of Figure 1. At first glance, the power-law $\psi(V)$ might seem to offer only minimal benefit over the linearized form. Both forms cross over, rather than asymptote, to the TFD curves. Also, the power-law curves, though reducing to a value of zero, as is required, do so with an infinite limiting slope, so that the limiting value of Γ is not positive, but remains zero. On the positive side, however,

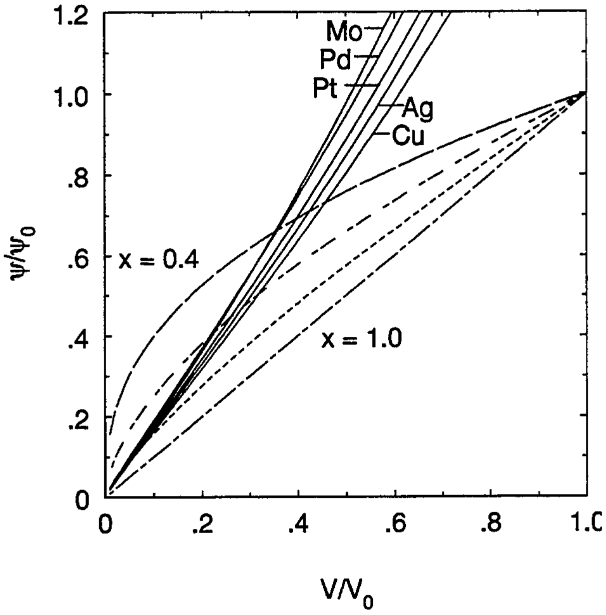

Figure 2. *Limiting ψ values for several metals (solid lines), obtained by inserting TFD cold-curve values into equation of state of Segletes. Superimposed (dashed lines) are limiting ψ values associated with the power-law ψ(V) assumption.*

the curve corresponding to an exponent of 0.8 does blend nicely into the TFD curves. And of the nine metals examined by Segletes [15,16], seven had values for $\Gamma_0\psi'$ (corresponding roughly to exponent x) in the 0.75 to 0.88 range, where the power law fit approximates the TFD asymptote very well.

As far as actual cold-compression data, the power law and the linear assumption for $\psi(V)$ are nearly identical for $\Gamma_0\psi'$ values of 0.8 over relative volumes from unity down to 0.5. In this region, the power law provides a slightly more concave-upward curvature to the cold curve, which can be mitigated by choosing a value of exponent x, slightly below the fitted value of $\Gamma_0\psi'$ (*e.g.*, whereas $\Gamma_0\psi'=0.76$ fits stainless steel, a value of $x=0.72$ fits comparably well). Over the range of available cold-compression data [10,21,22], the power-law and linear $\psi(V)$ formulations can be made virtually indistinguishable with appropriate selection of x, both resulting in excellent fits to the data.

On the expansion end of the curve, the power-law $\psi(V)$ is seen to provide an improved correlation to data. At high values of anharmonicity (~9), the linear model was able to match the universal cold-curve (UCC) fit of Rose *et al.* [10] with a value of $\Gamma_0\psi'$ approaching unity. The power law does equally well here, since for an exponent approaching unity, the linear and power forms are identical. However, for low anharmonicities (~3), no single value of $\Gamma_0\psi'$ could produce a match to the UCC model [15,16], but instead required a slope that was initially around 0.8, reducing to 0.55 at the larger expansions. Figure 3 compares the lattice potential energy predicted by the UCC model of Rose to that using the linear-ψ approximation. In this graph, the abscissa, a, represents the nondimensional, relative lattice spacing employed by Rose *et al.* [10].

By contrast, the power law $\psi(V)$ is able to match the UCC model closely with a single value of exponent x for a given anharmonicity. The value of the exponent x, which will provide a match to the UCC model, is given approximately by

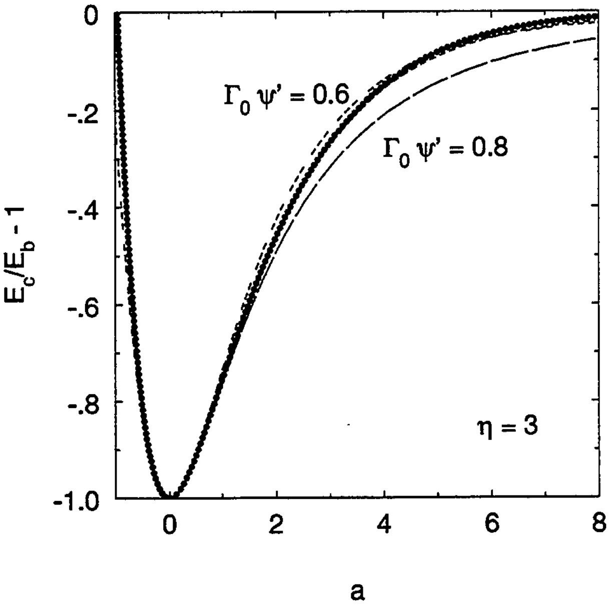

Figure 3. *Lattice binding energy as a function of nondimensionalized lattice spacing for a material. Dark line is UCC model of Rose et al. [10] Dashed lines are for model of Segletes [15,16], with linear* ψ *assumption, for two particular values of* $\Gamma_0\psi'$ *and an anharmonicity of 3.*

$$x = \frac{2}{3} + \frac{1}{3}\cdot(\eta/10) \quad , \tag{8}$$

where η is the value of anharmonicity, as defined by Rose *et al.* [10]. The magnitude of anharmonicity provides a metric on the deviation of an actual crystal's behavior from the harmonic idealization, in which the binding force of the atomic lattice would be proportional to the atom's displacement from the rest position. It is later shown that Segletes' model [15,16] approximates the harmonic solid described by Dugdale and MacDonald [5] when the value of exponent x is selected as 2/3, in accordance with the lower limit of eqn. (8), as $\eta \to 0$. Figure 4 compares the lattice binding energy predicted by the UCC model to that of the current model, when a power-law $\psi(V)$ is employed, the exponent being selected in accordance with eqn. (8). Though not labeled, the dark curve represents the UCC model, while the other five, essentially overlapping, curves are for the current model with anharmonicities of 2, 4, 6, 8, and 10.

4. GRÜNEISEN MODELS

Certain mathematical consequences of a power-law $\psi(V)$, eqn. (7), which are subsequently used, are easily derived and are simply stated as follows:

$$(\Gamma_0 \psi')_{V_0} = x \quad ; \tag{9}$$

$$\psi' = x \cdot (\psi/V) \quad ; \tag{10}$$

$$\psi' - n\cdot(\psi/V) = (x-n)\cdot(\psi/V) \quad ; \text{ and} \tag{11}$$

$$\psi\,[\psi'' - n\cdot(\psi/V)'] = (x-n)\,(x-1)\cdot(\psi/V)^2 \quad . \tag{12}$$

We now intend to show the primary result of this paper—namely, that three historical Grüneisen formulations (those of Slater [4], Dugdale and MacDonald [5], and the free-volume

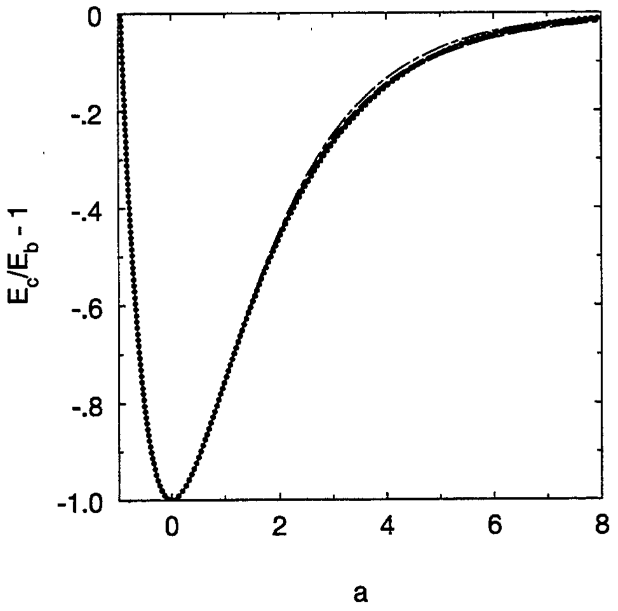

Figure 4. *Lattice binding energy as a function of nondimensionalized lattice spacing for a material. Dark line is UCC model of Rose et al. [10]. Dashed lines (essentially coincident) are for model of Segletes [15,16], with power-law ψ(V) assumption, for anharmonicity values of 2, 4, 6, 8, and 10.*

theory [6]) may be recovered through manipulation of the current model, if the material is idealized and higher-order terms are neglected. Start with eqn. (4) and multiply by a power of specific volume, V^n, where n is, at this point, still an arbitrary constant:

$$(p_c V^n) = \frac{E_b K^2 V^n}{\psi} (\Theta/\Theta_0)^K \ln(\Theta/\Theta_0) \ . \tag{13}$$

The first and second derivatives of this equation may be obtained in a tedious but straightforward manner as:

$$(p_c V^n)' = -\frac{E_b K^2 V^n}{\psi^2} (\Theta/\Theta_0)^K \left\{ [K + \psi' - n(\psi/V)] \ln(\Theta/\Theta_0) + 1 \right\} \tag{14}$$

and

$$\begin{aligned}(p_c V^n)'' = \frac{E_b K^2 V^n}{\psi^3} (\Theta/\Theta_0)^K \Big(&[K + 2\psi' - n(\psi/V)] \left\{ [K + \psi' - n(\psi/V)] \ln(\Theta/\Theta_0) + 1 \right\} + \\ &[K + \psi' - n(\psi/V)] - \psi[\psi'' - n(\psi/V)'] \ln(\Theta/\Theta_0) \Big)\end{aligned} \ . \tag{15}$$

Take eqn. (15) multiplied by $(-\psi)$ and divide by eqn. (14) to obtain:

$$-\psi \frac{(p_c V^n)''}{(p_c V^n)'} = 2K + 3\psi' - 2n(\psi/V) - \frac{\left\{ [K + \psi' - n(\psi/V)]^2 + \psi\, [\psi'' - n(\psi/V)'] \right\} \ln(\Theta/\Theta_0)}{1 + [K + \psi' - n(\psi/V)] \ln(\Theta/\Theta_0)} \ . \tag{16}$$

Eqns. (10–12) may be substituted into this expression to eliminate ψ', the left hand ψ may be broken out as V/Γ, and the equation may be algebraically manipulated to give

$$-\frac{3x-2n}{2}-\frac{V}{2}\frac{(p_cV^n)''}{(p_cV^n)'}=\Gamma\times\left(K-\frac{1}{2}\frac{\{[K+(x-n)(\psi/V)]^2+(x-n)(x-1)(\psi/V)^2\}\ln(\Theta/\Theta_0)}{1+[K+(x-n)(\psi/V)]\ln(\Theta/\Theta_0)}\right)\ . \tag{17}$$

The large term in parentheses at the end of this equation takes on the value K at ambient conditions and diminishes with compression as the logarithm terms exert their influence. The material parameter K, for materials studied by Segletes [15,16], has generally been close to the idealized value of unity [whereupon $(C_0/\Gamma_0)^2=E_b$]. For the sake of discussion, therefore, consider the large term in parentheses as unity, as an approximation to the situation near ambient volume [$\ln(\Theta/\Theta_0)\approx 0$] for an idealized material ($K\approx 1$). Eqn. (17) becomes

$$\Gamma=-\frac{3x-2n}{2}-\frac{V}{2}\frac{(p_cV^n)''}{(p_cV^n)'}\ . \tag{18}$$

In this equation, x, which denotes the power of $\psi(V)$, is a material parameter. If we select the value of parameter n, which has been arbitrary to this point, to be related to x in the following way:

$$n=3x-4/3\ , \tag{19}$$

then x may be eliminated from eqn. (18) to yield

$$\Gamma=-\frac{4-3n}{6}-\frac{V}{2}\frac{(p_cV^n)''}{(p_cV^n)'}\ . \tag{20}$$

Eqn. (20) is precisely the generalized result presented by Vaschenko and Zubarev [6] for the

Grüneisen function. In this equation, if $n=0$ (*i.e.*, when $x=4/9$), Slater's [4] result is obtained. If $n=2/3$ (*i.e.*, when $x=2/3$), then the result of Dugdale and MacDonald [5] is recovered. Finally, for the case where $n=4/3$ (*i.e.*, when $x=8/9$), the free-volume theory [6] expression follows. What this result tells us is that any one of the three historical Grüneisen models may be nominally applicable, depending on the properties of the material of interest. In the case of those metals already studied by Segletes [15,16], values for $\Gamma_0\psi'$, which correspond to exponent x at ambient conditions [via eqn. (9)], fell as low as 0.43 ($\approx$4/9) for molybdenum and as high as 0.88 ($\approx$8/9) for rubidium, with the vast majority of materials in the 0.75–0.85 range.

5. NONIDEAL EFFECTS

Let us now consider the practical influence of the large term in parentheses in eqn. (17), idealized as unity in the previous section. The leading K term in the braces affects the ambient value of the Grüneisen parameter, while the remaining term involving logarithms governs the rate at which the Grüneisen function changes with volume. If Segletes' equation of state is accepted, then the fact that the large term in parentheses diminishes with volume indicates that all of the historical Grüneisen formulations (Slater, Dugdale-MacDonald, and free-volume theory) will tend to overestimate the rate of change of the Grüneisen function with compression when using a cold-compression curve as the baseline.

Whereas the historical Grüneisen models are based on various theories of lattice vibration, the model of Segletes [15,16], in contrast, is shown to fit actual cold and Hugoniot data well to several megabars of pressure. Thus, if one accepts the validity of the Grüneisen assumption [$\Gamma = f(V)$], then, over the compression range of actual thermodynamic data to which Segletes' model fits well, one must accept as more accurate the mildly varying Γ of that model to the more widely varying estimates produced by the theories of Slater, Dugdale-MacDonald and/or the free-volume theory.

To see how the models compare, Figures 5–7 depict the Grüneisen functional behavior for a hypothetical material with parameter $K=1$ and $\Gamma_0=2$. These three figures depict four curves

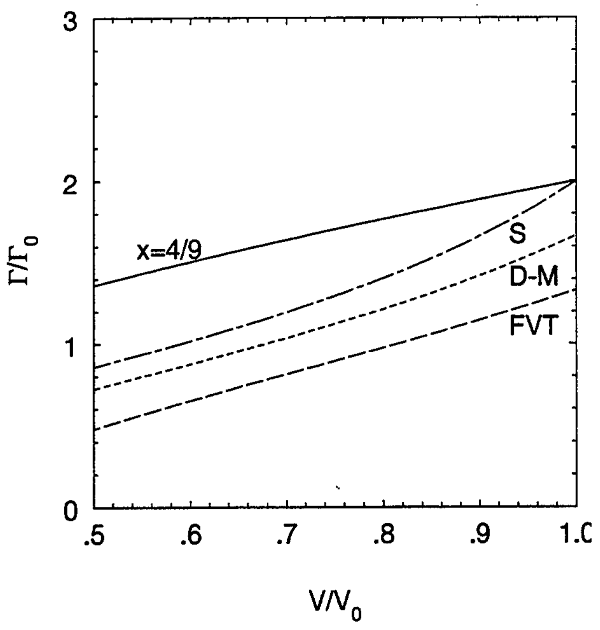

Figure 5. *Grüneisen function versus relative volume for hypothetical material with $\Gamma_0=2$ and $K=1$, using power-law $\psi(V)$, with $x=4/9$. Predictions using models of Slater [4], Dugdale and MacDonald [5], and the free-volume theory [6] are shown for comparison. Note that ambient value associated with power-law $\psi(V)$ matches that of Slater.*

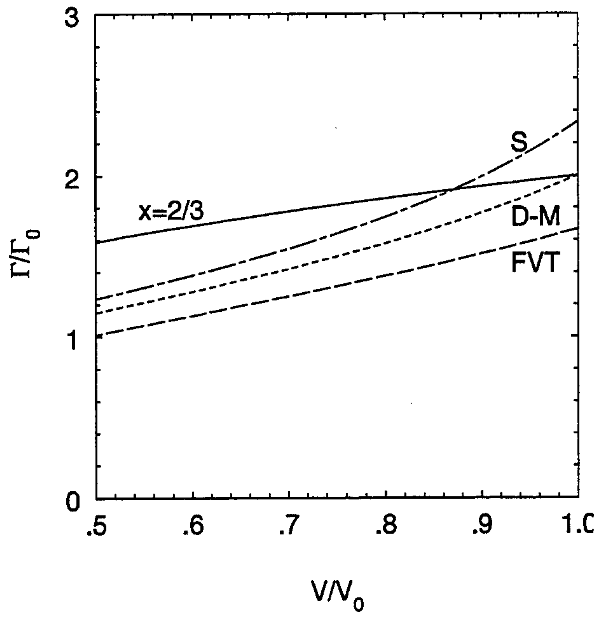

Figure 6. *Grüneisen function versus relative volume for hypothetical material with $\Gamma_0=2$ and $K=1$, using power-law $\psi(V)$, with $x=2/3$. Predictions using models of Slater [4], Dugdale and MacDonald [5], and the free-volume theory [6] are shown for comparison. Note that ambient value associated with power-law $\psi(V)$ matches that of Dugdale and MacDonald.*

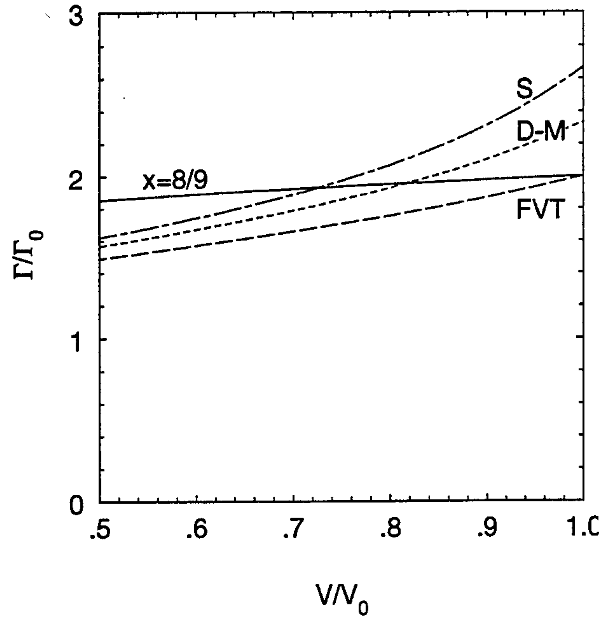

Figure 7. *Grüneisen function versus relative volume for hypothetical material with $\Gamma_0 = 2$ and $K=1$, using power-law $\psi(V)$, with $x=8/9$. Predictions using models of Slater [4], Dugdale and MacDonald [5], and the free-volume theory [6] are shown for comparison. Note that ambient value associated with power-law $\psi(V)$ matches that of the free-volume theory.*

each. In each graph, there is a Grüneisen curve corresponding to the power-law $\psi(V)$, with exponent x equal to 4/9, 2/3, and 8/9 respectively. The resulting cold curve from the current model is used to show what the models of Slater, Dugdale-MacDonald, and the free-volume theory would predict for the behavior of the Grüneisen function, depicted in each of the figures by curves labeled S, D-M, and FVT, respectively. Per eqn. (17), with K equal to unity, the ambient values for the current thory match the value of Slater when x=4/9, that of Dugdale and MacDonald when x=2/3, and that of the free-volume theory when x=8/9.

To see a comparison of the other models to that of Segletes, for an actual material, we consider the case of stainless steel, because of the availability of good cold-curve data out to 4 Mbar. Figure 8 shows the cold curve and Hugoniot for the steel. The data points are the same as those originally analyzed by Segletes [15,16]. However, unlike the earlier work, in which a linear $\psi(V)$ function was fit to the data with a slope of 0.76, the cold curve is fit on this occasion with a power law, with an exponent, x, of 0.72. The Hugoniot fit is excellent as far as data are available (out to 2 Mbar). If the Grüneisen assumption is accepted, then a power law exponent of 0.72 must provide an accurate reflection of the Grüneisen function down to a relative volume of 0.68 (where Hugoniot data stop), and most likely provides an accurate reflection of the function down to a relative volume of 0.55 (where cold data stop). Figure 9 shows the Grüneisen function corresponding to this fit with the solid curve. Overlaid on the figure are the results of the models of Slater, Dugdale and MacDonald, and the free-volume theory, which are derived from the curvature of the stainless steel cold-curve fit. As pointed out previously, the other theories all show a Grüneisen function that decreases more rapidly in compression than the current model and data suggest.

Of course, at very high compressions and temperatures, electronic and non-Grüneisen thermal effects will come into play, and so even if a simple power law for $\psi(V)$ might blend into the TFD cold curve, it will likely fail to capture the high-temperature thermal effects properly. For example, the shock Hugoniot of most materials asymptotes at a V/V_0 value of around 0.2. Such an asymptote corresponds to a relative compression, μ, of 4. Segletes [9] proved that the asymptotic value of the Grüneisen parameter on the Hugoniot cannot exceed (and, in fact, must

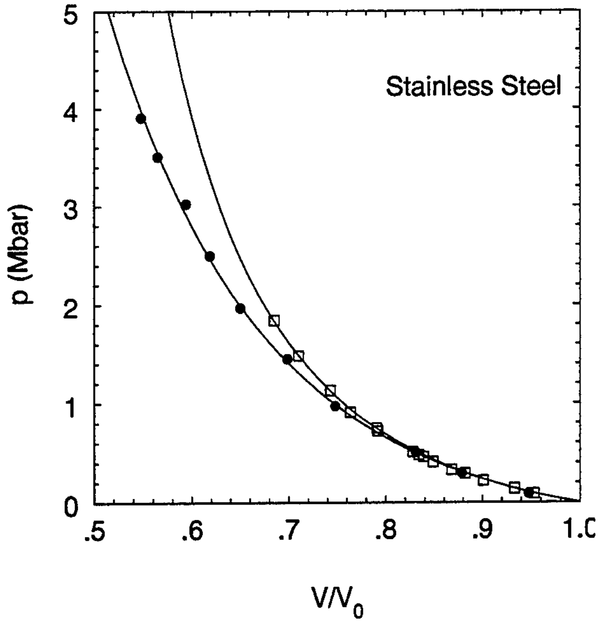

Figure 8. *Pressure-volume curves for stainless steel. Cold data to 4 Mbar shown with filled symbols. Hugoniot data to 2 Mbar shown with open symbols. Curves represent cold-compression and shock-Hugoniot curves with* Γ*=1.81 and power-law* $\psi(V)$ *exponent x, equal to 0.72.*

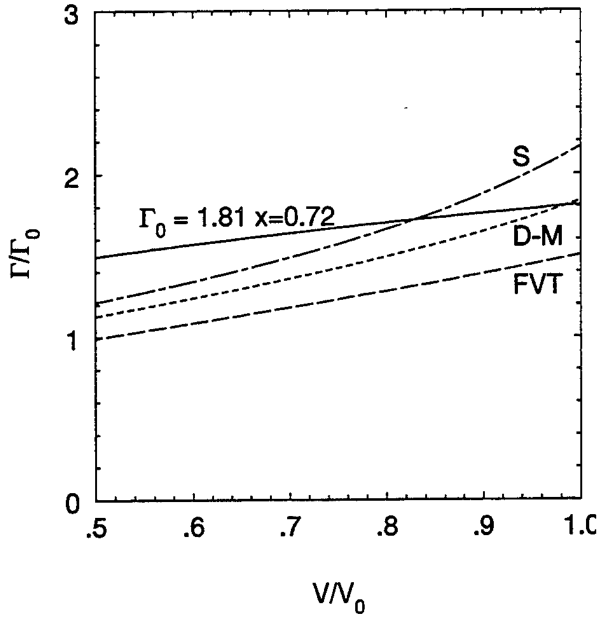

Figure 9. *Grüneisen function versus relative volume for stainless steel with* Γ_0 *=1.81, using power-law* $\psi(V)$*, with x=0.72. Predictions using models of Slater [4], Dugdale and MacDonald [5], and the free-volume theory [6] are shown for comparison.*

take on) a value of $2/\mu_{asym}$, which in this case, gives a value of 0.5. If the ambient value of the Grüneisen parameter were a typical value of Γ_0=2, then this asymptotic shock limit would possess a ψ/ψ_0 value of 0.8 at a V/V_0 value of 0.2. A quick look at the power-law curves of Figure 2 reveals that power-law $\psi(V)$ functions with exponents between 0.4 and 1.0 will not reach the point (0.2, 0.8). Thus, it would seem that the only way the Hugoniot can get to the point (0.2, 0.8) while the cold curve asymptotes to the TFD curves is if ψ is temperature dependent.

There is some reason to suspect such temperature-dependent behavior, by looking, for example, at the aluminum Hugoniot fit of Segletes [15,16]. When using the known ambient Grüneisen value (Segletes used 2.39) with the current model, the Hugoniot prediction was quite poor. But by using an ambient value of half that amount, which doubles the value of ψ, the aluminum Hugoniot and cold curve were fit with precision to 9 Mbar (V/V_0 = 0.4). One might be led to conclude that ψ is, indeed, temperature dependent and that, in the case of aluminum, a dependence occurs at relatively low temperatures. Such a thermal dependency would be able to explain how the Hugoniot can achieve relatively large asymptotic values of ψ/ψ_0, while, at the same time, the cold-curve value diminishes below this level. A significant thermal dependency of the Grüneisen function for aluminum has been reported by Grodzka [1]. In particular, he cites experimental work with shocked aluminum foam that shows the ambient-volume aluminum Grüneisen parameter to decrease from a cold value of 2.5 to a value below 1.4 at 6500 J/g of specific internal energy. Using a value of $3R/W$ to approximate the specific heat over this range (where R is the universal constant, and W is the atomic weight of aluminum), 6500 J/g corresponds to a temperature of approximately 7000 K, and would result from a moderate-strength shock in the 1.5 Mbar range.

As a final note, another way in which Segletes' model and those of Slater, Dugdale and MacDonald, and the free-volume theory may be contrasted is accomplished by rearranging eqn. (20) as

$$\left[2 + \frac{4-3n}{3}(\psi/V)\right]\psi\,(p_c V^n)' + \psi^2 (p_c V^n)'' = 0 \quad . \tag{21}$$

As before, the value of n determines which of the three older models is being discussed. This equation is first-order in $(p_c V^n)'$. By comparison, the current model, with a power-law $\psi(V)$ function, satisfies the following identity:

$$K^2 (p_c V^x) + \left[2K + x(\psi/V)\right] \psi (p_c V^x)' + \psi^2 (p_c V^x)'' = 0 \quad , \tag{22}$$

which is second-order in $p_c V^x$. These two equations are obviously different, in that eqn. (21) contains only derivatives of $p_c V^n$, while eqn. (22) involves both $p_c V^x$ and its derivatives. However, there is also a great deal of similarity in that not only are both equations hyperbolic, but for the idealized, harmonic case of $K = 1$ and $x = n = 2/3$, eqns. (21) and (22) differ only by the first term of eqn. (22).

If eqn. (19) is used to convert eqn. (21) from n-based to x-based (to consider cases when $n \neq x$), it can be shown (for K=1) that the transformed eqn. (21) also differs from eqn. (22) only in the first term of eqn. (22), regardless of the value of x. It is for for this reason that at ambient conditions, when p_c [and thus the first term of eqn. (22)] is zero, the value of Γ_0 from Segletes' model matches the older theories as a function of x, respectively. We can thus think of the older theories as each being applicable for a particular class of material (defined by material parameter x), but at the same time neglecting the $p_c V^x$ term necessary to capture the empirical data correctly.

6. CONCLUSIONS

In this report, the recently proposed equation of state of Segletes [15,16] has been examined for compatibility with prior theory. In the original work, an excellent match to available data and first-principles computations was achieved with the use of an assumed-linear expression for the $\psi(V)$ function, defined as V/Γ. In the current work, the use of a power law for $\psi(V)$ was examined in lieu of the linear relationship.

A number of positive outcomes resulted with the use of this power law. First, the low-volume limit for $\psi(V)$ approaches zero, which was shown to be desirable in order to have the current theory blend smoothly into Thomas-Fermi-Dirac cold-compression theory. Secondly, over the range of available cold-compression data, the linear and power-law fits can be made virtually indistinguishable, both providing excellent fits to the data. Finally, the binding energy function using a power-law $\psi(V)$ assumption was able to reproduce the fit of Rose *et al.* [10] better, especially in the area of lattice expansion. In this regard, an expression given by eqn. (8) shows a simple relation between material anharmonicity and the fitted parameter of the current model, in order to match the results of Rose *et al.* closely.

The primary result of the paper has been in showing how Segletes' model with a power-law $\psi(V)$, for the case of an idealized material, ignoring higher-order terms, is able to reproduce three historical Grüneisen function models, depending on the value of the fitted material parameter in Segletes' model. In particular, when the model parameter, which relates to the decrease of the Grüneisen function with compression, takes on a value of 4/9, the result of Slater [4] is obtained. When the parameter is taken as 2/3, the model of Dugdale and MacDonald [5] is recovered. Finally, if the model parameter takes the value of 8/9, the free-volume theory [6] expression follows.

The paper then has shown how Segletes' model differs from the historical Grüneisen models, if the higher-order terms are retained. It becomes apparent that the historical models of the Grüneisen-function predict a more rapid decrease of the Grüneisen function with compression than does the current model. Some speculations have been put forward here on the possibility of a temperature-dependent ψ function, which would, by definition, violate the Grüneisen hypothesis. Not only would this possibility help to explain some seeming-incompatibilities between the behaviors of the high-compression cold curve and the asymptotic Hugoniot, but a temperature-dependent ψ could also explain the otherwise anomalous behavior for aluminum cited by Segletes [15,16] in the relative volume range of 0.85 to unity. Finally, the older Grüneisen models and Segletes' model have been mathematically expressed in a different form, which clearly shows the latter's distinction from the others. In the case of the comparison between an idealized, harmonic material in the current model to the model of Dugdale and MacDonald, the distinction originates from a single $pV^{2/3}$ term, which represents a higher-order departure from the earlier theories.

There are several future efforts in this area that would be of utility. From a position of theoretical interest, it would be useful to reconcile the current theory with the very-high-pressure, equation-of-state theories, in something more than an empirical blend. Barring this reconciliation, however, the current theory has still shown its applicability into the megabar regime with excellent results, and should be of utility to those modeling material deformation resulting from impact and conventional explosive events in the ordnance regime. Also, the area of non-Grüneisen (and/or phase change) effects seems an interesting and important area for further work. For many materials, the Grüneisen approach seems perfectly adequate over a large range of compressions and pressures. However, there do seem to be materials (aluminum has been mentioned in this paper) for which non-Grüneisen effects at moderate pressures might provide an adequate explanation.

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A recently proposed equation of state is further examined for compatibility with prior theory. It is found, by adopting a more realistic functional relationship for one of the thermodynamic state variables and eliminating higher-order terms from the model, that three historical theories for determination of the Grüneisen function fall directly out of the recently proposed model. One is led to conclude that the newer model captures the essence of all of the older theories, each of which being applicable under an appropriate circumstance.

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