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# Phenomenological Hugoniot curves for transition metals up to 1 TPa

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## Abstract

We study the volume dependence of the Hugoniot pressure obtained from shock-wave experiments by using the tight-binding (TB) potential. We observe that the Hugoniot P-V curve of one metal resembles an effective cold P-V curve of the same element but having a stiffer repulsive term in the TB potential. We show that our ansatz is in systematic good agreement with experimental data for 12 close-packed metals. This observation allows us to derive a phenomenological relation between the Hugoniot curve and the 293 K isotherm. Using finite strain theory, this relation is extended to metals having non-close-packed structures.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

In shock physics experiments, a curve known as the Hugoniot curve is a valuable tool for analyzing a material's equation of state (EoS) [1]. If a solid with a defined initial pressure, density, and energy is subjected to a series of compression experiments of varying shock strengths, a set of new compression states can be plotted. The resulting (pressure versus volume) curve is the material's Hugoniot curve. Every material has a unique Hugoniot curve, which means that along the Hugoniot curve the temperature only depends on the volume and the initial solid state. From shock-wave (SW) experiments, extremely high pressures up to 1000 GPa can be reached in the solid. The shock wave travels at velocity  $u_s$  through the solid, accelerates its atoms from rest to velocity  $u_p$  and changes its density, atomic volume, pressure, and internal energy per atom from  $\rho_0$ ,  $V_0$ ,  $P_0$  and  $E_0$  to  $\rho$ , V,  $P_H$  and E. Assuming thermal equilibrium before and after the shock, these quantities must satisfy the Rankine– Hugoniot relations, derived from considerations of mass, momentum, and energy conservation:  $P_H - P_0 = \rho_0 u_s u_p$ ,  $V/V_0 = (u_s - u_p)/u_s$ , and  $(P_H + P_0)(V_0 - V) = 2(E - E_0)$ .

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Experimentally the Hugoniot curve ( $P_H$  versus V) is obtained from the measured quantities  $u_s$  and  $u_p$ , while by solving the latter equation together with a theoretical EoS, which relates P, V, and E, one can predict the Hugoniot curve [1]. Electronic structure calculations from first-principles theory now make possible an accurate estimation of the Helmholtz free energy and the resulting Hugoniot curve by taking into account the contributions of both phonons and the thermal excitation of electrons [2–4] (see also [5] for a detailed analysis of this elaborate method that accounts for possible phase changes along the Hugoniot curve).

In this paper we report a general property of the Hugoniot curves for transition metals. Our model relies on the phenomenological observation that a Hugoniot P-V curve is very similar to a low-temperature isotherm P-V curve of the same element but having an effective stiffer interatomic repulsion. Indeed, one can notice that for most metals the difference between the Hugoniot pressure and the isothermal pressure (both obtained at 293 K) tends to zero when the volume is close to  $V_0$  and increases when the volume decreases [1]. Since, a stiffer repulsion of the interatomic potential leads to very similar behaviors of the resulting pressure in these compression states, one can wonder whether an effective cold P-V curve could mimic the Hugoniot curve. Thus, and following this line of thought, our starting point in this work is an ansatz that consists in modeling the Hugoniot curve from an effective 'cold' E-V curve.

For this purpose, we assume an effective energy expression from a modified form of the tight-binding second-moment approximation (TB-SMA) potential developed in the past to describe structural properties of fcc transition metals [6, 7]. The proposed effective energy does not require any additional parameters and keeps the values of the usual four TB-SMA parameters unchanged. A comparison of the resulting effective P-V curve to experimental Hugoniot data shows a systematic good agreement for fcc Au, Ni, Pt, Rh, Pd, Ir, Ag, Cu, Pb, Al metals and hcp Cd, Zn elements. We conclude that the position of the Hugoniot curve with respect to the room-temperature isotherm is mainly driven by a common analytical relation that depends only on the equilibrium quantities:  $V_0$ , cohesive energy  $E_c$ , isothermal bulk modulus  $B_0$  and its pressure derivative  $B'_0$ . Then, we extend our description to bcc Mo, W, and Ta metals by combining the main properties of our model and finite strain theory. Finally, using classical thermodynamics relations, this work allows us to propose an analytical form of the temperature increase along the Hugoniot curve.

## 2. Results

We use the *N*-body potential formulated in the TB-SMA scheme. Restricting the interaction to the *Z* first neighbor atoms located at equivalent distance *r*, the bulk energy  $E_{\text{SMA}}$  per atom is written as [6]

$$E_{\rm SMA}(V) = AZe^{-px} - \xi\sqrt{Z}e^{-qx},\tag{1}$$

where  $x = r/r_0 - 1 = (V/V_0)^{1/3} - 1$ ,  $r_0$  is the first neighbor equilibrium distance and A, p,  $\xi$ , and q are the parameters of the model. From the experimental values of the cohesive energy  $E_c$ , atomic volume  $V_0$ , and the isothermal bulk modulus  $B_0$ , one can derive the following relations:  $\xi = pE_c/[(p-q)\sqrt{Z}]$ ,  $A = qE_c/[(p-q)Z]$  and  $pq = 9B_0V_0/E_c$  for cubic structures [6]. The fourth relation that fixes the values of the SMA parameters can be obtained from the shear elastic constants [8], from Rose's universal equation of state [9] where p/q = 2.95 [10] or from  $B'_0$  the pressure derivative of  $B_0$  where  $p + q = 3(B'_0 - 1)$ . In this study, the SMA parameters are taken from the original work of Rosato *et al* [8] for Ni, Cu, Ag, Pt, and Au elements and determined from the p/q = 2.95 relation for the other metals (see table 1). From equation (1) the volume dependence of the SMA pressure at T = 0 K is given by the following

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**Table 1.** The parameters of the model are taken from [8] for the X\* elements, otherwise they are calculated from the experimental values of  $r_0$ ,  $E_c$ , and  $B_0$  [21] and the p/q = 2.95 relation [10]. From equations (1) and (3) the resulting values of  $B_0/B_{\text{eff}}$ ,  $B'_0/B'_{\text{eff}}$ , and  $B''_0/B''_{\text{eff}}$  ratios are also reported.

	Ni*	Cu*	Ag*	Rh	Pd	Ir	Pt*	Au*	Pb	Al	Cd	Zn
<i>r</i> <sub>0</sub> (Å)	2.49	2.55	2.89	2.69	2.75	2.72	2.77	2.88	3.50	2.86	3.12	2.78
$E_{\rm c}~({\rm eV})$	4.44	3.50	2.96	5.75	3.94	6.93	5.86	3.78	2.03	3.39	1.16	1.35
р	10.00	10.08	10.12	10.19	10.71	10.74	10.80	10.15	10.08	7.75	12.01	10.89
q	2.70	2.56	3.37	3.65	3.58	3.64	3.50	4.13	3.42	2.63	4.07	3.69
$B_0$	196	137	105	278	183	341	262	166	41	74	47	63
$B'_0$	5.23	5.21	5.50	5.61	5.76	5.79	5.77	5.76	5.50	4.46	6.36	5.86
$B_0/B_{\rm eff}$	0.936	0.938	0.931	0.929	0.934	0.934	0.936	0.923	0.930	0.911	0.941	0.935
$B_0'/B_{\rm eff}'$	0.927	0.928	0.927	0.927	0.930	0.930	0.931	0.925	0.927	0.910	0.936	0.931
$B_0''/B_{\rm eff}''$	1.063	1.066	1.054	1.050	1.052	1.051	1.053	1.046	1.053	1.062	1.048	1.051

equation of state [8]:

$$P_{\rm SMA}(V) = \frac{3B_0}{(1+x)^2} \frac{e^{-px} - e^{-qx}}{p-q}.$$
(2)

As shown in figure 1, the TB-SMA model (equation (2)) predicts well the isothermal EoS obtained from static diamond-anvil-cell compression experiments. For certain elements (Cu, Pb, and Al), we also report 293 K isotherms obtained from *ab initio* calculations [2, 4] in order to show that the EoS from the TB-SMA model is accurate even at very high pressure.

We now propose an effective cold energy  $E_{\text{eff}}(V)$  to mimic the Hugoniot curves. This effective bulk energy per atom as a function of the volume is

$$E_{\rm eff}(V) = AZ(1+x)^{-\frac{p}{2}} e^{-\frac{p}{2}x} - \xi \sqrt{Z} e^{-qx},$$
(3)

where  $x = r/r_0 - 1 = (V/V_0)^{1/3} - 1$ . In this formulation we have only replaced the empirical exponential Born–Mayer term  $e^{-px}$  by a stiffer repulsive one of the form  $(1 + x)^{-p/2}e^{-px/2}$ . Note that at  $V = V_0$ ,  $E_{\text{eff}}(V_0)$  is minimum and is equal to the cohesive energy  $E_c$ . The effective bulk modulus calculated from our model (equation (3)) is stronger by a factor  $B_{\text{eff}}/B_0 = 1 + 1/[2(p-q)]$ . Its first pressure derivative  $B'_{\text{eff}}$  is also higher than  $B'_0$  according to the relation  $B'_0/B'_{\text{eff}} = [2(p-q)+1](p+q+3)/[9p-6q+2(p^2-q^2)+5]$ .

Assuming again that the Hugoniot curve could be represented from the cold  $E_{\text{eff}}(V)$  curve of equation (3), following this logic the volume dependence of the effective pressure is simply  $P_{\text{eff}}(V) = -dE_{\text{eff}}/dV$  and becomes

$$P_{\rm eff}(V) = \frac{3B_0}{(p-q)(1+x)^2} \left[ \frac{2+x}{2(1+x)} (1+x)^{-\frac{p}{2}} e^{-\frac{p}{2}x} - e^{-qx} \right].$$
 (4)

In figure 1, we report the effective pressure  $P_{\text{eff}}(V)$  deduced from equation (4) and the experimental Hugoniot curves for the 12 metals. A remarkable quantitative agreement is observed between our model and the Hugoniot experiments over a wide range of densities and pressures. For all metals considered in figure 1, the volume dependence of the Hugoniot pressure  $P_{\text{H}}(V)$  can be written as

$$P_{\rm H}(V) \approx P_{\rm eff}(V).$$
 (5)

Besides, our model shows that the Hugoniot pressure is essentially driven by the roomtemperature EoS (i.e.  $P_{\text{SMA}}(V)$ ) and the equilibrium quantities  $V_0$ ,  $E_c$ ,  $B_0$  and  $B'_0$  through the same phenomenological relation. By combining equations (2) and (4) and considering that x is small, we find that

$$\frac{\Delta P_{\rm eff}(V)}{P_{\rm SMA}(V)} \approx \frac{x(px-2)}{4(1+x)(1-e^{(p-q)x})},\tag{6}$$



**Figure 1.** The pressure–volume equation of state from equation (2) (solid red line) and Hugoniot curve estimated from equation (4) (dotted blue line). Parameters p and q are taken from table 1. Experimental static compression measurements (St.), shock-wave data (Sh.), and the 293 K *ab initio* isotherm (Th.) are also reported for comparison.

where  $\Delta P_{\text{eff}} = P_{\text{eff}} - P_{\text{SMA}}$ ,  $p = 3(B'_0 - 1)(1 + \lambda)/2$ ,  $p - q = 3\lambda(B'_0 - 1)$  and  $\lambda^2 = 1 - 4V_0B_0/(B'_0 - 1)^2/E_c$ .

We now wonder if the general Hugoniot property found for close-packed structure persists for bcc structure metals. Here, it should be recalled that the TB-SMA is unreliable for treating



**Figure 2.** The pressure-volume EoS (solid red line) obtained by combining finite strain theory (equation (8)) and 293 K *ab initio* isotherms [4]. For Mo,  $B_0 = 260$  GPa,  $B'_0 = 3.89$ ,  $B''_0 = -0.015$  GPa<sup>-1</sup>, for W,  $B_0 = 302$  GPa,  $B'_0 = 4.00$ ,  $B''_0 = -0.013$  GPa<sup>-1</sup>, and for Ta,  $B_0 = 189$  GPa,  $B'_0 = 3.70$ ,  $B''_0 = -0.016$  GPa<sup>-1</sup>. The Hugoniot curve (dotted blue line) is obtained by replacing the  $B_0$ ,  $B'_0$ , and  $B''_0$  terms in equation (8) by  $B_{\text{eff}}$ ,  $B'_{\text{eff}}$ , and  $B''_{\text{eff}}$ , respectively, using equation (7).

bcc structures and that the attractive part of the cohesive energy of non-close-packed structures requires a more detailed description of the electron density of states (above the second-moment approximation) [6, 7]. Another TB-SMA limitation can be seen from the experimental values of the  $\lambda^2$  term, which is negative for Mo, W, and Ta bcc elements giving no solution for the (p, q) couple. However, since the effective energy proposed in this work in equation (3) differs by the SMA bulk energy from the repulsive term only, we believe that our modeling of the Hugoniot curve can be extended to bcc metals. To prove this assumption, we note that a strong signature of our model concerns the values of the effective bulk modulus  $B_{\text{eff}}$  and its pressure derivatives  $B'_{\text{eff}}$  and  $B''_{\text{eff}}$ . Indeed, systematic ratios are observed in table 1, and for most closepacked metals we find that

$$B_0/B_{\rm eff} \approx B_0'/B_{\rm eff}' \approx 0.93, \qquad B_0''/B_{\rm eff}'' \approx 1.05.$$
 (7)

Thus, the Hugoniot curve can be seen as a cold isotherm where the  $B_0$ ,  $B'_0$ , and  $B''_0$  terms are replaced by  $B_{\text{eff}}$ ,  $B'_{\text{eff}}$ , and  $B''_{\text{eff}}$ , respectively. To test this viewpoint, we use the Eulerian finite strain theory. In this formulation, the pressure as a function of the Eulerian finite strain variable  $f = [(V/V_0)^{-2/3} - 1]/2$  is a polynomial in strain [11]

$$P = B_0[1 + a_1f + a_2f^2 + \cdots][3f(1 + 2f)^{2.5}],$$
(8)

where  $a_1 = 3(B'_0 - 4)/2$  and  $a_2 = 3[B_0B''_0 + B'_0(B'_0 - 7) + 143/9]/2$  are the third- and fourth-order terms, respectively [11]. In figure 2, by fitting the finite strain EoS (equation (8)) to recent *ab initio* EoS, we obtain the  $B_0$ ,  $B'_0$ , and  $B''_0$  values for Mo, W, and Ta elements. Then combining equations (7) and (8), we also plot in figure 2 the resulting Hugoniot and compare it to experimental data. If at high pressure and for tantalum the Hugoniot pressure is underestimated, the agreement between shock-wave experiments and our model is almost perfect for Mo and W metals, indicating that the phenomenological relation found in our work also applies for some bcc metals.

Finally, it is interesting to note that from our derivation of both the isothermal pressure  $P_{\text{SMA}}(V, T_0)$  in equation (2) and the Hugoniot pressure  $P_{\text{eff}}(V, T_{\text{H}})$  in equation (4), it is possible to estimate the increase of the temperature  $T_{\text{H}}$  along the Hugoniot curve that only depends on volume. Indeed, by using the classical formulation of the thermal pressure [3, 12], the Hugoniot pressure can be written as:

$$P_{\rm H}(V, T_{\rm H}) = P_0(V, T_0) + \Delta P_{\rm th}(V, T_{\rm H}), \tag{9}$$

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<b>Table 2.</b> Values of parameters $a$ , $\gamma_{\theta}$ , and $p_{\theta}$ used in equation (15).											
	Cu	Ag	Pt	Au	Pb	Al					
$a (\text{GPa } \text{K}^{-1})$ $\beta_e (10^{-3} \text{ J mol}^{-1} \text{ K}^2)$ $\gamma_e$	0.0070 0.695 0.91	0.0058 0.646 1.18	0.0073 6.8 2.4	0.0072 0.729 1.6	0.003 55 2.98 1.7	0.0053 1.35 1.61					

**Table 2.** Values of parameters *a*,  $\gamma_e$ , and  $\beta_e$  used in equation (13).

where  $T_0 = 293$  K. According to equations (4) and (5), we have shown that the thermal pressure  $\Delta P_{\rm th}(V, T_{\rm H})$  can be roughly written as

$$\Delta P_{\rm th}(V, T_{\rm H}) \approx P_{\rm eff}(V, T_{\rm H}) - P_{\rm SMA}(V, T_0) = \Delta P_{\rm eff}(V). \tag{10}$$

At high temperature (roughly above the Debye temperature) the thermal pressure of a transition metal can be separated into a lattice and an electronic contribution:

$$\Delta P_{\rm th}(V, T_{\rm H}) \approx \frac{\gamma_{\rm l}(V)}{V} E_{\rm th}^{\rm l}(V, T_{\rm H}) + \frac{\gamma_{\rm e}(V)}{V} E_{\rm th}^{\rm e}(V, T_{\rm H}), \qquad (11)$$

where  $\gamma_{\rm I}(V)$  and  $\gamma_{\rm e}(V)$  are the lattice and an electronic Grüneisen parameters. In the high temperature approximation, the thermal lattice energy  $E_{\rm th}^{\rm l} = 3k_{\rm B}T$  while the thermal electronic energy  $E_{\rm th}^{\rm e} = 1/2\beta_e(V/V_0)^{\gamma_e}T^2$  depends on volume with  $\beta_e$  defined as the coefficient of electronic heat capacity that is proportional to the density of electron states at the Fermi level. The value of  $\gamma_{\rm I}(V)/V$  is estimated from the high temperature approximation  $\gamma_{\rm I}(V)/V = a/3k_{\rm B}$  [12] where  $a = \alpha_V B^{\rm T}$  is the average product of the thermal expansion coefficient  $\alpha_V$  and the isothermal bulk modulus  $B^{\rm T}$ .

Consequently, we obtain from equations (10) and (11) a simple polynomial form of the Hugoniot temperature  $T_{\rm H}$ ,

$$\Delta P_{\rm eff}(V) = a T_{\rm H}(V) + \frac{1}{2V_0} \gamma_{\rm e} \beta_e (V/V_0)^{\gamma_{\rm e} - 1} T_{\rm H}(V)^2, \qquad (12)$$

with the analytical solution

$$T_{\rm H}(V) = \frac{V_0}{\gamma_{\rm e}\beta_e} (V/V_0)^{1-\gamma_{\rm e}} \bigg[ -a + \sqrt{a^2 - \frac{2}{V_0}\gamma_{\rm e}\beta_e (V/V_0)^{\gamma_{\rm e}-1}\Delta P_{\rm eff}(V)} \bigg].$$
(13)

Finally, to illustrate how the temperature increases along the Hugoniot curve, we plot for a few cases in figure 3 the Hugoniot temperature as a function of the relative volume  $(V/V_0)$  according to equation (13). Some experimental and calculated data obtained by other authors are also reported for comparison. The values of the parameters a,  $\gamma_e$ , and  $\beta_e$  used in equation (13) are collected in table 2. Estimation of the parameter a requires the knowledge of high temperature thermal expansion  $\alpha_V$  and isothermal bulk modulus  $B^T$ . In practice the thermal expansion is directly measured and the isothermal bulk modulus is deduced from direct measurements of the adiabatic bulk modulus  $B^S$  by using the relation  $B^T = B^S [1 + T V \alpha_V^2 B^S / C_p]^{-1}$  [13]. Data for  $B^S$  are taken from [14] (apart from those for Pt for which [15] is used). Data for heat capacities at constant pressure  $C_p$  are taken from [16] and the  $\alpha_V$  values are obtained from [17]. Finally for  $\gamma_e$  and  $\beta_e$ , [18] and [19] are used, respectively.

## 3. Conclusions and discussions

Using phenomenological arguments, we propose an empirical relation to predict the Hugoniot P-V curve for metals. This relation is verified with success from experimental data for 15



**Figure 3.** Estimation of the temperature along the Hugoniot curve (dotted blue line) using equation (13). Experimental measurements (Exp.) and calculated temperatures (Calc.) are also reported for comparison.

metals having fcc, hcp, and bcc structures. Future efforts should focus on establishing the underlying physics of the proposed P(V) relation. However, the effective approach used in this work allows us to drawn the following important conclusion: many metals obey to a common relation between the Hugoniot P-V curve and the room-temperature isotherm. This relation depends essentially on equilibrium quantities.

To conclude, we should again insist on the phenomenological character of the approach chosen in this work. In this paper we do not claim that the repulsive interactions are changed in the shocked state nor that the Hugoniot pressure is the volume derivative of the Hugoniot energy. We simply observe that the Hugoniot curve looks very similar to an effective cold E(V) curve of the same element having a stiffer repulsion. Following the logic of this ansatz the effective pressure that mimics the Hugoniot is seen as the volume derivative of the effective cold E(V) curve (equation (4)). Such phenomenological approaches are frequent in physics and materials science. For instance in the field of 'driven alloys' [20], where alloys are maintained in nonequilibrium conditions by some external dynamical forcing (e.g. irradiation, ball milling), one uses an effective free energy functional to describe the so-called 'dynamical equilibrium states' of driven alloys and assess their relative stability. This effective free energy functional is a Lyapunov function of the kinetic problem, which does not have all the properties of a free energy. Another example that is directly related to the topic discussed in this work, is the 'Universal features of the equation of state of metals' by Rose et al [9]. The analytical EoS function assumed by these authors is purely phenomenological. This universal relation has been the subject of great debate. It has been tested from *ab initio* calculations and from experimental data. Some physical insights into the nature of this binding energy-distance relations were given by [10]. This 'Universal empirical curve' was very useful, in particular for

the parametrization of interatomic potentials similar to the TB-SMA potential. More generally, in most cases this type of ansatz has been found to be at the origin of important methodological developments identifying and quantifying the underlying physics. Finally, let us add that since the TB-SMA potential has been applied with success in the past to alloys [7], it would be interesting to extend and test this phenomenological approach to estimate the Hugoniot curves for alloys with a definite composition.

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