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Critical analysis of universality at shock pressures

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A critical analysis of the meaning of universality at shock pressures is presented, and a careful comparison is made of previous work by the present authors and other approaches to universality. To illustrate some relevant points in the discussion we shall consider in particular an interesting approach recently proposed to obtain universal relationships describing the high-pressure behavior of matter. The approximations made, the basic equations, the different kinds of scaled variables, and the various types of plots currently used are some of the points thoroughly discussed and compared in different approaches to universality.

I. INTRODUCTION

Shock waves in solids, liquids, and gases have been widely studied, during the last four decades from both the experimental and the theoretical points of view. The primary data obtained in shock experiments are in general shock and particle velocities. These are converted, using the Rankine-Hugoniot conservation equations, into thermodynamic (pressure-volume) data. Regardless of the type of representation used, the data for different materials seem to be completely unrelated. A principle of order was introduced with the so-called linear relationship between shock and particle velocities. This relationship, firmly based on experimental grounds, seems to be valid for all sorts of materials: pure elements, compounds, mixtures, solids, liquids, and gases, and is now widely accepted and used in the field of high-pressure physics.

We have taken advantage of this linear relationship to formulate a system-independent thermodynamics of high pressures. The obtaining of an analytical expression for the temperature along the shock-compression Hugoniot curve led us to establish a high-pressure equation of state.[1] Furthermore, by the use of a law of corresponding states for materials at shock pressure,[2] we succeeded in obtaining system-independent equations for the Hugoniot temperature, for the linear relationship between shock and particle velocities, and for an equation of state at high pressures.[2,3] So, what seemed to be completely unrelated data can be described and represented by universal equations and curves, valid for all materials.[4]

It is the purpose of the present paper to make a critical analysis of universality at shock pressures, and to make a careful comparison of our work with other approaches to universality.[5–7] We shall consider in particular, as an example to illustrate some relevant points of the discussion, the interesting approach recently proposed by Shively, Stein, and Robertson.[5]

II. CORRESPONDING STATES AND UNIVERSALITY

In dealing with shock compression, one must be careful in defining the various concepts, which are sometimes misunderstood. The Hugoniot is the curve representing states of shock compression of a given material. The direct experimental data on the Hugoniot are in general measured values of shock and particle velocities. These can be transformed, through the use of the Rankine-Hugoniot conservation equations of mass and momentum, into pressure-volume or pressure-particle velocity data. It should be clearly understood that the Hugoniot does not represent equation-of-state data, because states of shock compression are not states of thermodynamic equilibrium. It is also commonplace to speak of pressure-volume data or of pressure-particle velocity data as "equation-of-state data." In general this is not true because the dependence on a thermal variable, temperature or energy, is lacking.

We have already mentioned the linear relationship between shock and particle velocities

$$U_s = A + BU_p \ , \tag{1}$$

where U_s stands for the shock velocity, U_p for the particle velocity, and A and B are two parameters characteristic of each material. This relationship is strongly supported by the experimental data on all sorts of materials; some attempts have also been made to explain this linear behavior as a consequence of the existence of a limiting volume V_L under shock compression.[8–12]

Another pertinent comment refers to the van der Waals equation in reduced variables. Sometimes the "law of corresponding states" used by van der Waals to obtain the scaling parameters that make it possible to represent his equation of state in universal form is referred to as *the* law of corresponding states. It is to be noted that, within the context of thermodynamics, it is possible in principle

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to find not only one law, but several different laws of corresponding states associated with different phenomena. For a thorough discussion of this point see for example the works of Ellis[13] in general, and the one of Prieto[2] concerning a high-pressure law of corresponding-states formulation.

To avoid mistakes and misunderstandings, it is necessary to examine carefully the definition and meaning of the term "corresponding states." Therefore, the following points ought to be considered:

(1) The states always refer to two different materials and they are always associated with a given phenomenon, for example shock compression.

(2) The states are always related to a given equation or set of equations involving two or more of the variables used to describe the behavior of matter in relation to the phenomenon under consideration and they concern its representation rather than the behavior of the two materials in relation to this phenomenon.

(3) If $1,2,3,\ldots,N$ different variables are involved in the description of the phenomenon under consideration by a given equation, then a set of N different parameters $Q_1,Q_2,\ldots,Q_N$ can be defined as the ratio of the values of each one of the variables for the two different materials.

(4) The two equations describing the phenomena of the two different materials are then considered, and in one of them the set of N equations defining the Q_i's is introduced. One obtains then two different equations describing the phenomenon for the same material. Since the two equations ought to be identical, this imposes conditions on the values of the set of N parameters Q_i. Thus, a set of N equations involving these parameters is obtained.

(5) If this set of simultaneous equations has a solution, then the set of ratios for the N different variables defines "corresponding states" of the two materials in relation to the phenomenon under consideration and to the equation describing this phenomenon.

(6) But if this is the case, then a set of N scaled or reduced variables describing this phenomenon can be defined as the ratio of each one of the variables to the Q_i related to the variable considered.

(7) As a consequence of this definition, for each one of the variables involved, the scaled values are the same for the two materials. That is, in any numerical or graphical representation of the phenomenon the two materials are represented by one and the same point.

(8) It is then said that the two materials are in corresponding states.

(9) A law of corresponding states can then be formulated, and a system-independent or universal equation describing the phenomenon under consideration can be obtained. It is in this context that expressions such as "universal Hugoniot" or "universal isotherm at high pressures" should be understood.

It becomes then quite clear that the behavior of matter in relation to a given phenomenon does not depend on whether corresponding states for two different materials can be defined. The behavior is determined by the material itself and cannot be changed by the use of any given representation for the phenomenon under consideration.

III. TWO APPROACHES TO UNIVERSALITY

Following the procedure just outlined, Prieto[2] succeeded in expressing the equation for the Hugoniot in universal form, either in the pressure-volume or in the pressure-particle velocity representations. Let ρ_0 be the initial density and ρ the density at pressure P. The scaled variables used within this formalism are then defined by

$$p=P(\rho_0 A^2/B)^{-1} \tag{2}$$

for pressure,

$$x=B[1-(\rho_0/\rho)] \tag{3}$$

for the relative compression,

$$u_s=U_s/A \tag{4}$$

for shock velocity, and

$$u_p=U_p/(A/B) \tag{5}$$

for particle velocity.

In terms of these scaled variables, the linear relationship takes the universal form

$$u_s=1+u_p\ . \tag{6}$$

This equation, being system independent, fits the thousands of experimental points accumulated up to now for hundreds of materials of many different kinds: pure elements, compounds, mixtures, solids, liquids, and gases. With this criterion, the universal equations for the Hugoniot

$$p=x(1-x)^{-2} \tag{7}$$

or

$$p=u_p(1+u_p) \tag{8}$$

fit the same experimental data in the pressure-volume and in the pressure-particle velocity representations, respectively.

It is to be clearly understood that the proof of the validity of an equation expressed in universal form depends completely on the fact that a law of corresponding states can be formulated for the phenomenon under consideration, or, alternatively, on the fact that the set of simultaneous equations for the Q_i's has a solution. Concerning the goodness of fit[14] of the equation or of its associated graphical representation, once a criterion such as the measure of "total square error" given by Mallows[15] is chosen, its measure might be different depending on whether physical or reduced variables are used, but this apparent discrepancy is irrelevant. Thus, an equation such as the one for the Hugoniot can be and is sometimes expressed using different scaling parameters, but the goodness of fit is exactly the same as that of the original equation once the variables are converted from the reduced set to the physical set of variables. The same remark can be made in relation to the values of the parameters used to scale the different variables. These values may of course be most important to determine how good or bad is the original equation in fitting the experimental data. But if those values are somehow used in scaling the variables, the goodness of fit does not change.

We have already insisted on the fact that an equation for the Hugoniot is not an equation of state by itself. However, it is possible to consider equilibrium states in the neighborhood of shock states and use the thermodynamics of equilibrium to correlate the variables describing these states. This is the procedure we followed first to formulate and solve the thermodynamic differential equation for the Hugoniot temperature, and, from this, to obtain a complete equation of state at high pressures.[1,4] The equation for the Hugoniot temperature can also be expressed in reduced variables, and it agrees reasonably well with the available experimental data on shock temperatures already reported by several authors, as well as with computed values obtained by numerical integration of the differential equation for the Hugoniot temperature.[16,17]

In a recent paper Shively, Stein, and Robertson[5] propose a different procedure to obtain a universal Hugoniot. They also use scaled variables for pressure, for shock velocity, and for particle velocity. Many references are made to the system-independent formalism for the thermodynamics of high pressures developed by the present authors, but rather they follow what we have called the dimensional analysis approach, without the support of a law of corresponding states.[18] Since the slope B of the linear relationship between velocities cannot be eliminated from the equation by this kind of scaling, they do not get a universal equation for the Hugoniot. They claim nevertheless that universality is achieved if one assumes that the slope B is a universal constant and consequently has the same value for all materials. This is an interesting conjecture that certainly deserves closer analysis.

Shively and collaborators use $\rho_0 A^2$ to scale pressure, and A to scale both velocities. This kind of scaling has been used sometimes by several authors,[19-21] and we have already shown[18] that it is equivalent to the scaling we use in our formalism i.e., $\rho_0 A^2/B$ for pressure, A for the shock velocity, and A/B for particle velocity, if the values of the parameter B are about the same for the materials under comparison.

What makes their approach most puzzling is that among the materials they use to support their conjecture the values of the parameter B go from 0.95 for silicon carbide to 1.85 for H_2O and yet they get seemingly acceptable goodness of fit to the experimental data for about 20 materials. We think that this is mainly due to the following: (a) they use an approximation valid only at low compression, and (b) they are using a pressure-particle velocity representation in a log-log plot. In the following we will examine closely these facts and their consequences.

The basic equation used by Shively and collaborators for their comparison with the experimental data and with our approach is the one obtained for the Hugoniot by the use of the Rankine-Hugoniot conservation equations complemented by the linear relationship between shock and particle velocities. This equation reads

$$p=\rho_0 U_p(A+BU_p) \ . \tag{9}$$

In their approach they use the scaled variables

$$p^*=p/(\rho_0 A^2) \tag{10}$$

for pressure, and

$$u_s^*=U_s/A \ , \quad u_p^*=U_p/A \tag{11}$$

for shock and particle velocities, respectively. So, the equation for the shock Hugoniot becomes

$$p^*=u_p^*(1+Bu_p^*) \ , \tag{12}$$

whereas for the linear relationship between velocities one gets

$$u_s^*=1+Bu_p^* \ . \tag{13}$$

These equations are to be compared with Eqs. (8) and (6) obtained within our formalism for pressure and for the linear relationship, respectively.

In making this comparison it is important to note that the variables used in the two formalism are related by the following equations:

$$p=Bp^* \ , \tag{14}$$

$$u_s=u_s^* \ , \quad u_p=Bu_p^* \ . \tag{15}$$

If small pressures are considered, and this is the approximation used by Shively, Stein, and Robertson, the term in U_p^2 in the three equations (8), (9), and (12) becomes negligible so that one gets linear equations in the particle velocity, with slope 1.0 in both approaches. Because of Eqs. (14) and (15), the difference between their representation and ours then amounts to a mere change of scale by a factor B for both the pressure and the particle-velocity axes.

Concerning the graphical representation in the U_s-U_p plane, and the comparison between both formalisms within this approximation, because of Eq. (13) the slopes of the straight lines are different for each material in Shively, Stein, and Robertson's formalism, whereas in ours the plot of Eq. (8) is one and the same straight line for all materials. In the pressure-particle velocity representation, and with the log-log plot they use, it is clear from Eqs. (14) and (15) that Eq. (8) becomes

$$\log_{10}p^*+\log_{10}B=\log_{10}u_p^*+\log_{10}B \ . \tag{16}$$

So, if both formalisms are compared using this plot, the representation is the same in both cases, a straight line with slope 1.0, but the data for a given material in our formalism are displaced with respect to the data for the same material in their formalism by an amount $\log_{10}B$ on both axes. Of course, as already mentioned, and within this approximation, the goodness of fit is the same in both cases, and is as good as the fit of the linear relationship to the experimental data on velocities.

We are now in the position to understand the remarkable agreement between the so-called master curve of Shively, Stein, and Robertson and the experimental data on shock compression for various materials having values of the slope B very different from the value 1.335 they proposed for this parameter. We shall first comment that the relationship between the normalized pressure defined by Eq. (10) and the normalized particle velocity defined in Eq. (1) should be linear, not because it seems to be so in the log-log plot, but because the quadratic term is being neglected. Consequently, the pressure vs particle velocity relationship becomes linear with slope exactly equal to 1.0. A second comment is that the correlation obtained by Shively, Stein, and Robertson is not fortuitous or due to the fact that the slope B of the linear relationship between velocities is the same for all materials. What happens is that the difference in the values of the slope B is masked by the log-log plot. In this representation, the data for a given material in their formalism become indistinguishable from the data for the same material in our universal formalism, since the net effect of differences in the value of B is a shift in the representation of the data along the same straight line. But this is only true at low compressions and as long as a log-log plot is used. In a different kind of plot, as we shall see later, the differences due to the different values of B become stronger.

IV. COMPARISON WITH EXPERIMENTAL RESULTS

In their approach to universality at shock pressures, Shively, Stein, and Robertson considered 12 pure elements and 10 compounds, one of these in the liquid phase. The range of values of the slope B, as determined by fitting the shock data by straight lines, goes from 0.950 for silicon carbide to 1.850 for water. To make the comparison between the two formalisms, we have selected only five of these materials such that they cover the whole range of values of B. Special consideration is given to this parameter because the main difference between the two formalisms lies here.

In our formalism B is included in the scaling factors used to reduce some of the variables. In their formalism it is concluded that B has the same value for all materials, and consequently that it cannot play a significant role in the formalism.

Table I shows the pertinent data for the five materials we selected to make the comparison. For each material the so-called shock parameters are first given: initial density ρ_0, and intercept A and slope B of the linear relationship between velocities, as defined by Eq. (1). The basic experimental data are given in the form of shock velocity $U_s = U_s^*$, particle velocity U_p(expt), and Hugoniot pressure P(expt). These quantities are not independent, but related by the equation

$$P = \rho_0 U_s U_p \; . \tag{17}$$

TABLE I. For each of the materials listed in the first column, ρ_0 is the initial density, and A and B are the coefficients of the linear relationship between velocities, Eq. (1). P_h(expt) expressed in kbar and U_p(expt) expressed in km/s are the experimental values of the Hugoniot pressure and the particle velocity, as reported in Ref. 22. p is the universal Hugoniot pressure, as defined by Eq. (8). p^* is the normalized pressure as defined by Eq. (12). u_s, u_p, u_s^*, and u_p^* are the universal and normalized shock and particle velocities as defined by Eqs. (4), (5), and (11), respectively.

Material	P(expt)	p	p^*	U_p(expt)	u_p	u_p^*	$u_s = u_s^*$
Copper	164	0.176	0.118	0.41	0.155	0.104	1.13
$\rho_0 = 8.930$ g/cm^3	320	0.344	0.231	0.73	0.276	0.185	1.25
$A = 3.940$ km/s	652	0.700	0.470	1.25	0.472	0.317	1.48
$B = 1.489$	1018	1.093	0.784	1.74	0.658	0.442	1.66
	1318	1.416	0.951	2.10	0.794	0.533	1.78
	1581	1.698	1.140	2.37	0.896	0.602	1.89
Lead	838	2.725	1.796	1.64	1.227	0.809	2.21
$\rho_0 = 11.34$ g/cm^3	1388	4.515	2.976	2.25	1.683	1.109	2.68
$A = 2.028$ km/s	1383	4.498	2.965	2.25	1.683	1.109	2.68
$B = 1.517$							
Water	38.5	3.124	1.689	1.110	1.360	0.735	2.30
$\rho_0 = 1.00$ g/cm^3	61	4.949	2.675	1.465	1.795	0.970	2.75
$A = 1.51$ km/s	103.5	8.398	4.539	1.980	2.426	1.312	3.46
$B = 1.85$	119	9.655	5.219	2.180	2.672	1.444	3.61
	138	11.197	6.052	2.365	2.898	1.567	3.86
Graphite	81	6.714	3.685	1.350	2.116	1.161	3.17
$\rho_0 = 1.628$ g/cm^3	120	9.946	5.459	1.739	2.726	1.496	3.65
$A = 1.162$ km/s	139	11.521	6.325	1.829	2.867	1.573	4.02
$B = 1.822$	200.5	16.618	9.121	2.331	3.654	2.005	4.55
	210.0	17.406	9.553	2.442	3.828	2.101	4.56
Silicon carbide	155	0.074	0.078	0.46	0.055	0.058	1.34
$\rho = 3.124$ g/cm^3	422	0.201	0.211	1.37	0.163	0.171	1.23
$A = 8.000$ km/s	677	0.322	0.339	2.16	0.257	0.270	1.25
$B = 0.950$	999	0.475	0.500	3.00	0.356	0.375	1.33
	1170	0.556	0.585	3.56	0.423	0.445	1.31

These basic experimental data are transformed to the values used in our formalism (p,u_s,u_p) through Eqs. (2), (4), and (5). The values used in Shively, Stein, and Robertson's formalism (p^*,u_s^*,u_p^*) were obtained through Eqs. (10) and (11).

Figure 1 shows a pressure vs particle velocity (p^*,u_p^*) plot of the Hugoniot compression curves for $B=1.00$, 1.50, and 1.85. These were obtained using Eq. (12). The experimental data for copper, lead, water, graphite, and silicon carbide listed in Table I are also included in this figure. In all cases the source of the experimental data is Van Thiel's compendium.[22] Regarding this figure it is very important to point out the following.

(a) It is a log-log plot, not a Cartesian representation.

(b) Because of Eqs. (8) and (12), a plot of the Hugoniot for $B=1.00$ coincides with the plot of the universal Hugoniot in our formalism.

(c) For low values of compression, let us say for u_p^* less than 0.05, the quadratic term in Eqs. (8) and (12) is negligible, and all the Hugoniot curves in this log-log plot collapse into one and the same straight line of slope 1.00,

$$\log_{10}p^*=\log_{10}u_p^* \ . \tag{18}$$

(d) The black and white blocks shown in the low-compression linear section of these plots illustrate the shift previously discussed in relation to Eq. (16). Suppose that shock data for a given material fall within the black area when expressed in Shively, Stein, and Robertson's variables (p^*,u_p^*). Because of Eq. (17) the same data would then fall in the white area when expressed in our variables (p,u_p). Within this approximation the same would happen for any other material with a different value of B. We shall now understand the apparent universality of the plots in Shively, Stein, and Robertson's work.

(e) If only the experimental data were shown in this figure, without any of the computed Hugoniot curves for different values of the parameter B, it would seem that all the experimental data can be fitted, to a very good approximation, by one single curve. This is how Shively, Stein, and Robertson reached the conclusion that one single Hugoniot, that of Eq. (12) with $B=1.335$, suffices to fit the experimental data on shock compression for all materials. They therefore conclude that B ought be considered a universal constant.

Before going farther in the analysis of the log-log representation, let us consider two different representations of the same Hugoniot curves. Figure 2 shows plots of the same Hugoniots and of the same experimental data shown in Fig. 1, the difference being that the representation is now made in Cartesian coordinates. Here the effect of the differences in the value of the parameter B becomes evident. For a given value of u_p^* the difference in p^* between the values for $B=0.95$ and 1.85 is about 30% or higher. If the value $B=1.335$ proposed in Ref. 5 is accepted, the differences in p^* would be smaller, but since they fall within the range 20–25 % anyhow, this is obviously unacceptable because the experimental error for these kinds of data is less than 1%.

The same comments can be made in relation to a Cartesian plot of the Hugoniots using shock and particle velocities as variables. Such a plot is shown in Fig. 3. Here again the same Hugoniots and experimental data of

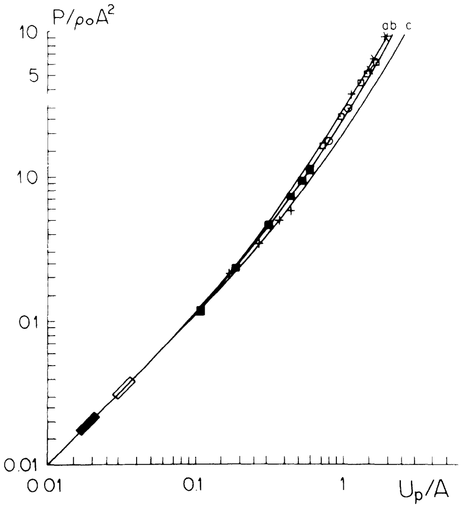

FIG. 1. Log-log plot of pressure vs particle velocity Hugoniots for different values of the parameter B: curve a, 1.85, curve b, 1.50, and curve c, 1.00. The last one coincides with our universal Hugoniot. The experimental data are those reported in Van Thiel's compendium (Ref. 22) normalized using Eqs. (10) and (11) for the materials: ■, Cu (1.489); ⊙, Pb (1.517); □, H_2O (1.850); ∗, graphite (1.822), and +, silicon carbide (0.950).

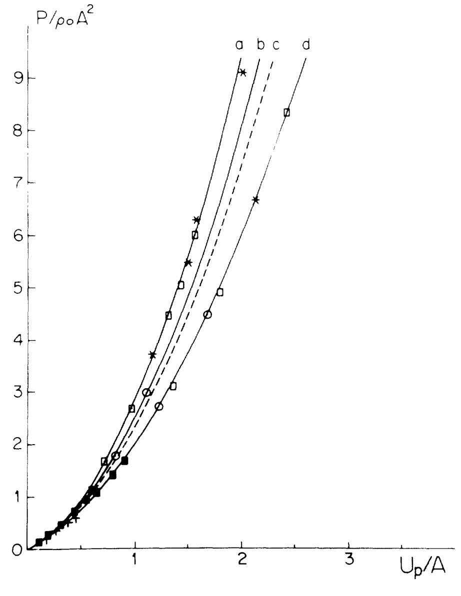

FIG. 2. Cartesian plot of pressure vs particle velocity Hugoniots for different values of the parameter B: curve a, (1.85), curve b, 1.50, and curve c, 1.335, as proposed by Shively, Stein, and Robertson (Ref. 5), and curve d, 1.00, which coincides with the universal Hugoniot. The experimental data are shown in both Shively, Stein, and Robertson's approach and in our universal approach, for the same materials of Fig. 1: ■, Cu (1.489); ⊙, Pb (1.517); □, H_2O (1.850); ∗, graphite (1.822), and +, silicon carbide (0.950).

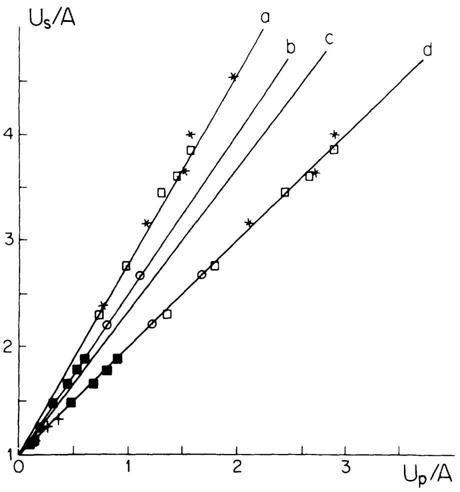

FIG. 3. Cartesian plot of the linear relationship between shock and particle velocities for different values of the slope B: curve a, 1.85, curve b, 1.50, and curve c, 1.335 as proposed by Shively, Stein, and Robertson (Ref. 5), and curve d, 1.00), which coincides with the universal linear relationship. The experimental data are shown in both Shively, Stein, and Robertson's formalism and in our universal formalism, for the same materials of Fig. 1: ■, Cu (1.489); ⊙, Pb (1.517); ⊡, H_2O (1.850); ∗, graphite (1.822), and +, silicon carbide (0.950).

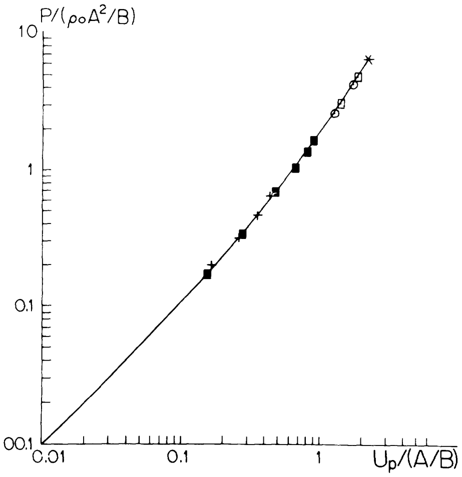

FIG. 4. Log-log plot of our universal Hugoniot, as computed using Eq. (8). Experimental data are shown for the same materials of Fig. 1: ■, Cu (1.489); ⊙, Pb (1.517); ⊡, H_2O (1.850); ∗, graphite (1.822), and +, silicon carbide (0.950).

Figs. 1 and 2 are reproduced. It becomes evident that differences between experimental values and values computed for any mean value of the slope, such as $B=1.335$, are too high, by about 20–25 %, and consequently unacceptable, since the experimental errors in these kinds of measurements are less than 1%. This figure shows also a straight line of slope 1.0. This is identical to a plot of the universal Hugoniot of Eq. (6) in our approach. It shows also the experimental data for the same materials used in this comparison, but transformed to the variables u_s and u_p we use. The agreement is as good as the linear approximation in fitting the experimental data on velocities.

Figure 4 shows finally a plot of the universal Hugoniot in our formalism, but using the log-log representation proposed by Shively, Stein, and Robertson. The variables used here are p and u_p. The solid curve is the universal Hugoniot computed using Eq. (8). The experimental data shown are also the same as in the previous figures. The agreement is excellent. This is of course a consequence of the validity of the law of corresponding states in general, independently of the material or of the kind of representation used.

V. FINAL REMARKS

We are thus inclined to think that the excellent agreement found by Shively, Stein, and Robertson in fitting the experimental data on shock compression with one single value of the slope B is circumstantial. It is a consequence of the use of a log-log plot for the (p^*, u_p^*) Hugoniots. In the best of the cases it can be considered as an acceptable approximation to the Hugoniot of other materials, but limited to this kind of representation, and it cannot be extended to the phenomenon of shock compression in general. It seems then that universality cannot be reached by numerical analysis. This situation contrasts strongly with universality at shock pressures as proposed in our approach as a consequence of the formulation of a law of corresponding states. This is valid in general, it is not an approximation, and it does not depend on the type of plot used to represent the phenomena under consideration.

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