

Shock adiabats for ultrahigh pressures

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The shock adiabats of 52 solids were calculated by using the Thomas-Fermi method with quantum and exchange corrections. According to the estimates, their accuracy is satisfactory at pressures exceeding ~ 50 Mbar. For quartz and five metals (Al, Fe, Cu, Cd, Pb), the calculated adiabats and the experimental shock-compression data are used to construct interpolation adiabats that describe well both the experimentally investigated region and the region of pressures up to thousands of megabars.

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1. INTRODUCTION

Explosions, high-velocity collisions, and other processes that produce large local energy concentration give rise to strong shock waves.^[1-3] The extremal states produced behind the wave front are described, in terms of the chosen thermodynamic variables, by the shock-compression curves (the Hugoniot adiabats^[1]). The adiabats in the condensed media were registered experimentally^[3-5] up to pressures 5-10 Mbar, were calculated by the Thomas-Fermi (TF) statistical model of the atom for pressures above 300 Mbar,^[6] and were obtained by interpolation in the intermediate region.^[7,8] The reliability of this interpolation is inadequate, since the accuracy of the TF model for weakly heated matter at $P \sim 300$ Mbar is low (it becomes satisfactory only at much higher pressures).

Corrections to the TF model, making the model more precise and extending the regions of its applicability, were proposed in^[9-11]. The Thomas-Fermi theory with quantum and exchange corrections (TFC) was used recently^[12-14] for numerical calculations of all the thermodynamic functions of a large number of substances. The availability of these calculations makes it possible to interpolate more reliably the shock adiabats between the experimental and theoretical regions, and to interpret anew the results of the individual relative-compressibility measurements^[7,8,15,16] carried out in recent years in the pressure range from 15 to 60 Mbar.

It is known^[1-5] that absolute measurements of the dynamic compressibility are performed in specially organized experiments, in which two kinematic parameters of the shock wave can be registered—its propagation velocity D and the mass velocity u of the material behind its front. From these quantities, using the conservation laws, one determines the pressures, densities, and internal-energy increments. In the comparative experiments mentioned above, the shock-wave velocities were registered only as they passed successively through layers of the investigated substances. The determination of the wave parameters in one of the layers was based on a solution of the problem of the decay of the discontinuity at the contact, and presupposes knowledge of the dynamic adiabat and of the equation of

state of the other layer. This method has yielded record high pressures for iron, copper, cadmium, and quartzite, but the interpretation of these experiments was based on "standard" adiabats of lead and aluminum, constructed by using the TF asymptotic form of the adiabat.

We present in this paper the principles and results of the calculation of shock adiabats on the basis of an improved TFC model and compare them with experiments at "low" pressures. On the basis of the new calculations and comparison-measurement data, we obtained for quartz and five metals (Al, Fe, Cu, Cd, and Pb), the adiabats characterizing their dynamic compressibility at low, high, and ultrahigh pressures. The analysis has made it possible to determine more accurately the range of parameters in which the TFC model can be effectively used to obtain the equations of state.

2. CALCULATION OF THE ADIABATS AND COMPARISON WITH EXPERIMENT

1. In the assumed model, just as in the Wigner-Seitz theory, the material is broken up into neutral atomic cells, each of which contains one nucleus and its surrounding electrons. The electron gas in the cell is described by the TFC model, which has been investigated in detail in^[9-11]. It considers a quasiclassical gas of electrons interacting with the nucleus and with one another. The zeroth approximation is the TF model, in which the density of the electrons in phase space is determined by the Fermi distribution at arbitrary temperature:

$$\rho(\mathbf{r}, \mathbf{p}) = 2[1 + \exp\{(E - \mu)/T\}]^{-1}, \quad E = \frac{1}{2}p^2 + V(\mathbf{r}), \quad (1)$$

where μ is the chemical potential. The intra-atomic potential $V(\mathbf{r})$ satisfies the Poisson equation

$$\nabla^2 V = +4\pi\rho(\mathbf{r}), \quad (\nabla V)_{\text{bound}} = 0, \quad \rho(\mathbf{r}) = \int \rho(\mathbf{r}, \mathbf{p}) d\mathbf{p}. \quad (2)$$

Two corrections to the energy (1) are then introduced to refine the TF model, namely the Dirac volume correction and the Weizsäcker quantum correction (with a modified coefficient). This makes Eqs. (1) and (2)

somewhat more complicated, but extends greatly the region in which the model is applicable. The solution of the corrected problem (1) and (2) yields the atomic potential, from which it is possible to calculate all the thermodynamic functions of the electron gas. Owing to its complexity, the problem can be solved only by numerical means.

The boundary condition of Eq. (2) is equivalent to the condition that the electron density remain smooth on going from one atomic cell to another. This condition takes effective account of the interaction between charged particles from neighboring cells, and incorporates this interaction in the potential energy of the cell. As a result, the cells themselves can be regarded as non-interacting and their thermodynamic potentials can be added.

It is also necessary to take into account the motion of a cell as a unit, this being due to the vibrations of the nuclei about the equilibrium position. We shall describe this motion with the aid of the perfect-gas model, a reasonable procedure at high temperature or at low density of the material. The motion of the cells has very little effect on the energy of the electron gas inside the cell, so that the total energy of the material is equal to

$$E = n[E_e(T, v) + E_{nuc}(T, v)] \approx n[E_e(T, v) + \frac{3}{2}kT]. \quad (3)$$

Here n is the total number of cells, while the subscripts "e" and "nuc" label the contributions of the electron gas and of the cell motion per cell. The remaining thermodynamic functions are constructed in similar fashion. Generally speaking, it is necessary to add to expression (3) the thermodynamic functions of the equilibrium radiation, which begins to contribute substantially at $T \geq 1$ keV.

For chemical compounds it is convenient to use the additional average-atom approximation: the different atoms are replaced by atoms of one sort, such that

$$\langle Z \rangle = \frac{1}{n} \sum_i n_i Z_i, \quad \langle A \rangle = \frac{1}{n} \sum_i n_i A_i. \quad (4)$$

The total number of nuclei and electrons is conserved here (Z need not be an integer).

The TFC model does not take into account the effect exerted by the shell structure of the atoms on the thermodynamics. The allowance for the thermal contribution of the nuclei, made in the ideal-gas approximation, is also much simpler in this model.

2. The shock adiabats describe a sequence of states behind the fronts of shock waves with different amplitudes. They are obtained by simultaneous solution of two equations:

$$\varepsilon(P, v) - \varepsilon_0 = \frac{1}{2} P (v_0 - v), \quad \varepsilon(P, v) = \varepsilon_{TFC}(P, v). \quad (5)$$

The first of them is the Hugoniot relation written for the case of motion of a shock wave through uncompressed material, while the second is the equation of state of the

TABLE I. Shock adiabats of solids in the TFC model.

Substance	a	b	10 ⁶ c	Substance	a	b	10 ⁶ c	Substance	a	b	10 ⁶ c
Li	2.02	1.233	276	Sr	1.51	1.177	77	Pr	2.41	1.175	62
Be	5.44	1.216	280	Y	2.37	1.185	72	Nd	2.42	1.175	60
Na	1.73	1.206	144	Zr	3.28	1.188	72	Sm	2.50	1.175	60
Mg	3.12	1.204	151	Nb	4.05	1.191	68	Eu	1.74	1.171	58
Al	4.14	1.203	154	Mo	4.38	1.186	85	Gd	2.50	1.175	59
K	1.15	1.183	122	Rh	5.47	1.184	87	Tb	2.59	1.175	58
Ca	1.84	1.191	107	Pd	5.19	1.184	85	Tu	2.76	1.174	57
Sc	2.97	1.200	98	Ag	4.52	1.185	74	Yb	2.05	1.170	56
Ti	4.01	1.198	108	Cd	3.48	1.188	60	Os	5.12	1.182	54
V	5.19	1.192	127	In	2.91	1.185	62	Pb	2.72	1.172	52
Cr	6.01	1.192	124	Su	2.80	1.184	62	SiO ₂	5.04	1.206	178
Mn	5.93	1.190	123	Sb	2.58	1.182	59	LiF	5.93	1.213	225
Fe	6.20	1.190	120	Te	2.26	1.179	62	NaCl	3.11	1.204	137
Co	6.59	1.187	129	Cs	0.78	1.159	68	KCl	2.29	1.201	103
Ni	6.67	1.189	116	Ba	1.34	1.169	59	KBr	2.22	1.188	96
Cu	6.15	1.190	107	La	2.19	1.176	61	CsBr	2.02	1.179	67
Zn	5.06	1.190	104	Ce	2.40	1.176	61	CsI	1.80	1.173	66
Ga	3.78	1.193	87								

material in accordance with the TFC model. The parameters in the Hugoniot equation are the true specific volume V_0 of the material under normal conditions and the initial experimental value of the internal energy $\varepsilon_0 = \varepsilon_0(0, v_0)$, which is equal to the negative binding energy (the energies are reckoned from the level of neutral atoms located at infinity). With this choice of v_0 and ε_0 , the difference between the TFC adiabat and the true adiabat is due only to the inaccuracy of the model in the region of the compressed-state parameters. With decreasing shock-wave amplitude, the error in the calculation of the dynamic pressure increases and becomes infinite at $v = v_0$, since $\varepsilon_{TFC}(0, v_0) \neq \varepsilon_0$.

The principal results of the numerical calculations of the shock adiabats can be presented in compact form by using kinematic variables. The dependence of the shock-wave velocity D on the mass velocity u is established from the values of P and v calculated from the conservation laws:

$$P = v_0^{-1} D u, \quad v = v_0 D^{-1} (D - u). \quad (6)$$

It turns out that the obtained functions $D(u)$ are approximated with good accuracy by the quadratic expressions

$$D = a + b u + c u^2. \quad (7)$$

The coefficients c are small here, so that the calculated functions are almost linear up to very high pressures.

The coefficients of (7) for a large number of solids are listed in Table I.¹⁾ At velocities u exceeding 15–20 km/sec, they are close to the calculated shock adiabats in the values of the density and pressure, with accuracy ~1%. Of course, the physical accuracy of this approximation does not exceed the accuracy of the model itself.

3. The calculated adiabats of several substances are compared with the results of absolute measurements of the dynamic compressibility.^[3, 5, 17–23] This is done in the form of $P(\sigma = \rho/\rho_0)$ plots in Fig. 1 and in the form of $D(u)$ plots in Fig. 2. Figure 1 shows for comparison the shock adiabats calculated with the electron gas described only by the TF model without corrections. It is seen that allowance for the quantum and exchange corrections alters significantly the lower section of the adiabat as well as the value of the maximum compression. Therefore the use of the TF adiabats as asymptotic representations of the experimental curves leads to large errors.

We note that the functions $D(u)$ are approximately linear also when the thermodynamics is described by the TF model.

As shown in Fig. 2, the experimental $D(u)$ plots are concave upward. The changes in the slopes are due to the increase, in the large-amplitude waves, of the fraction of the thermal pressure with simultaneous decrease of the thermal elasticity of the nuclei with temperature. At pressures 5–10 Mbar the derivatives of the adiabats approach the derivatives of TFC curves. Since the calculation does not reflect the individual electron structure of the elements, the TFC adiabats, just as the zero isotherms, lie either above or below the experimental ones. A comparison of calculation with experiment for other metals and ionic compounds leads to analogous results.

On the whole, the discrepancy between calculation and experiment at pressures ~ 10 Mbar is quite large. At identical pressures, the discrepancy between the density curves reaches 8–10%. Thus, at the limit of the experimental range, the accuracy of the calculations by the the TFC model is insufficient and the model can be used only to obtain correct asymptotic values. For a number of substances, however, there is accidental cancellation of the physical errors of the calculation of the thermodynamic functions of the nuclei and the electrons. Examples of such substances are, in particular, iron and quartz, as seen from Figs. 1 and 2.

3. THE ADIABATS OF QUARTZ AND OF FIVE METALS

1. Interpolation curves of any variant should be tangent to the experimental adiabats and should merge at certain but presently unknown pressures with the TFC adiabats. The latter, as shown above, have in this case realistic slopes when the calculations are extended to

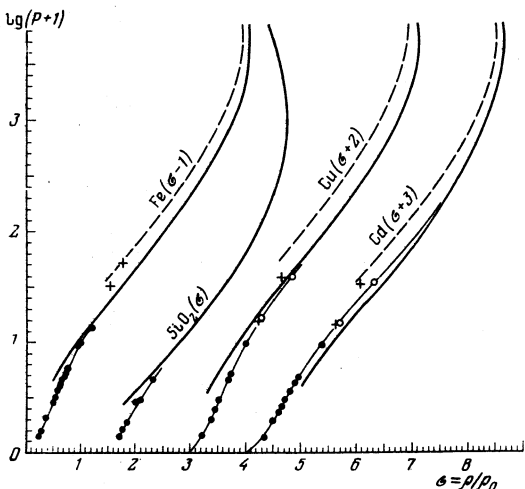


FIG. 1. Shock adiabats $P(\sigma)$ of quartz, iron, copper, and cadmium. The solid thick lines are the results of calculation by the TFC model, the thin lines are interpolation curves, and the dashed lines are calculations by the TF model. ●—Absolute measurements in shock waves, ○—new interpretation of the comparative measurements, +—old interpretation of the comparative measurements.

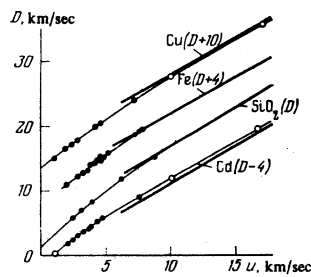


FIG. 2. $D-u$ diagram of copper, iron, quartz, and cadmium. Thin lines—results of absolute (●) and relative (○) measurements (see Table III below); thick lines—adiabats in accordance with the TFC theory.

low pressures, and do not pass very far from the upper experimental points.

For substances in which the discrepancy between the calculation and experiment is particularly small as a result of cancellation of the errors, the “linear $D-u$ interpolation” is single-valued and sufficiently reliable. It is based on choosing slightly modified coefficients of formula (7), such that a curve fits well the TFC adiabat at pressures $P > 300$ Mbar, and is tangent to the upper section of the experimental curve. All that is usually necessary for this purpose is to alter slightly the coefficients in Table I. Since the coefficient c remains small as before, the interpolation is practically linear in $D-u$ coordinates. By applying the described procedure to quartz and iron, we obtain corrected interpolation adiabats; next, using the data of [7, 8, 15, 16] on the comparative compressibility, we obtain the adiabats of aluminum, lead, copper, and cadmium.

2. *Adiabats of quartz and iron.* The results of absolute measurements of the dynamic compression of quartz [23, 18] and iron [3, 5, 17, 18, 21, 22] are approximated by parabolas of the form (7), but with a negative third term. The coefficients of the trinomials (obtained for SiO_2 in [24]) are obtained by statistical least-squares reduction of the entire available experimental information. The numerical values of the coefficients for the experimental ranges and the limits of the ranges are given in Table II. The same table gives the parabolic interpolation expressions for the upper branches. They differ from those of Table I only in that the first coefficients are slightly larger—by 0.35 km/sec for iron and 0.1 km/sec for quartz. The positions of the obtained curves on Fig. 1 are almost indistinguishable from the TFC adiabats.

3. *Adiabats of aluminum and lead.* The Hugoniot

TABLE II.

Substance	$D(u)$, km/sec	Range
SiO_2	$1.21+2.02u-0.044u^2$	$0 < u < 9.2$
	$4.95+1.206u+0.000178u^2$	$9.2 < u$
Fe	$3.79+1.717u-0.0251u^2$	$1.4 < u < 10.43$
	$6.54+1.19u+0.00012u^2$	$10.43 < u$
Al	$5.35+1.364u-0.0119u^2+0.000186u^3$	$0 < u < 35$
	$4.19+1.203u+0.000154u^2$	$35 < u$
Pb	$2.01+1.544u-0.0363u^2+0.00099u^3$	$0 < u < 17.05$
	$2.72+1.172u+0.000052u^2$	$17.05 < u$
Cu	$3.93+1.496u$	$0 < u < 3.7$
	$3.56+1.898u-0.0273u^2$	$3.7 < u < 9.01$
	$5.78+1.205u+0.000075u^2$	$9.01 < u$
Cd	$2.40+1.774u-0.049u^2$	$0 < u < 6.08$
	$4.22+1.177u+0.000085u^2$	$6.08 < u$

TABLE III.

Screen			Sample				
Substance	D_{exp} , km/sec	u , km/sec	Substance	D_{exp} , km/sec	u , km/sec	P , Mbar	$\sigma = \rho/\rho_0$
SiO ₂	13.60	7.16	Al	14.53	6.92	2.73	1.910
	15.28	8.55		15.96	8.34	3.61	2.094
	18.43	11.16		18.72	11.00	5.58	2.425
	19.92	12.39		20.05	12.26	6.66	2.573
	33.00	23.18		32.55	23.11	20.38	3.449
Fe	25.70	16.08	Pb	20.72	15.26	35.86	3.795
	31.90	21.27		26.12	20.10	59.55	4.341
Pb	14.64	9.84	Cu	17.82	9.98	15.89	2.274
	22.17	16.58		26.18	17.01	39.76	2.854
Cu	17.47	9.70	Cd	16.08	10.09	14.01	2.682
	25.30	16.18		23.88	16.67	34.39	3.312

curves of quartz and iron, used as standards, make it possible, by using the procedure of the reflection method,^[1-5] to obtain the points on the adiabats of aluminum and lead up to pressures 20 and 60 Mbar from the data of the experiments described in^[6] and^[7,16]. The initial values of the measured wave velocities D_{exp} in quartz or iron screens placed in front of the samples, and in the samples themselves, are given in Table III. The same table lists the remaining characteristics of the shock waves in the screens and in the samples. The values of u_{SiO_2} and u_{Fe} were obtained directly from D_{SiO_2} and D_{Fe} by using formula (7) with the coefficients of Table II. The parameters of the compressed Al and Pb samples were obtained by graphic solution of the shock-decay problem. The solutions were obtained using different variants of the equations of state of SiO₂ and Fe. The obtained values correspond to a Grüneisen coefficient $\gamma = v(\partial P/\partial \epsilon)_v = 0.55$. Variation of this coefficient in the range 0.3-0.8 changed the mass velocities in lead by $\pm 0.7\%$ and did not affect the parameters of the shock waves in aluminum.

The results of the calculations are shown graphically in a semilogarithmic scale in Fig. 3. The information obtained in this manner for the dynamic compressibility of aluminum covers the range from 3 to 20 Mbar. The upper point lies close to the theoretical TFC adiabat of aluminum. The entire dynamic compression curve of aluminum is described by two analytic expressions from Table II: a cubic expression up to 43 Mbar and a quadratic expression with the coefficients of Table I for higher pressures.

In the case of lead, its two upper points at $P = 40$ and 60 Mbar lie, within the limits of experimental error, on the TFC adiabat (Table I). At lower pressures, its dynamic compressibility curve is approximated by a cubic equation, just as in the case of aluminum.

The obtained shock-compression curves of aluminum and lead differ strongly from the previously published interpolations.^[7,8] At 50 Mbar, the density difference is 4.9% for aluminum and 10.7% for lead.

4. Adiabats of copper and cadmium. A similar interpretation of the experiments of^[15,16] on the comparative compressibility of lead, copper, and cadmium has made it possible to obtain in succession the adiabat of copper from the adiabat of lead, and then the shock-compression curve of cadmium from the adiabat of copper. The positions of the figurative points on the adiabats of copper

and cadmium are shown in Table III, and the approximating expressions for the different ranges are shown in Table II. The compression curves are plotted in Fig. 1. The upper branches of adiabats of copper and cadmium pass through the high points and satisfy simultaneously the linear $D-u$ interpolations.

4. LIMITS OF APPLICABILITY OF THE TFC MODEL

The main shortcoming of the TFC model is the averaged description of the electron shells (bands in the case of dense matter), without a detailed account of their structure. The theoretical estimates of the role of the shells are complicated and unreliable. Reasonable estimates are obtained by comparing the model with a large group of experiments.

By way of the criterion for the applicability of the TFC model we arbitrarily choose the 5% error level in the determination of the density from the given pressure and temperature. An analysis of the experimental curves of the atomic volumes $v(Z)$ at fixed pressures has shown in this case for zero temperature^[10] that the model is valid at $P \geq 300$ Mbar. The physical cause of the applicability is in this case the broadening of the electron band with increasing density of matter, which leads to a decrease in the role of the shell effects.

It is seen from Figs. 1 and 3 that on the shock adiabats the experimental results (more accurately, the interpolation adiabat) agrees with the TFC model within the limits of the indicated accuracy already at pressures $P \sim 50$ Mbar. The latter is explained by the fact that shock waves of such amplitude heat the matter to $T > 10$ eV, and the increase of the temperature smoothes out the shell effects even more strongly than an increase in the density.

The approximate limit of the applicability of the TFC model is shown in Fig. 4; the lower section of the limit is drawn on the basis of a comparison with plasma characteristics^[25] and calculations based on the equations of a weakly-ideal plasma under ionization equilibrium.^[26] The figure indicates also the temperature and pressure regions that can be attained in experiment at the present time. It is seen that the TFC model cannot be used at moderate pressures or temperatures, but supplements and replaces well the experiment in the

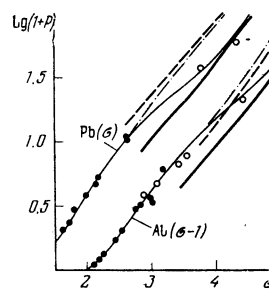


FIG. 3. Adiabats $P(\sigma)$ of lead and aluminum. The symbols are the same as in Fig. 2. The thin lines are the interpolation adiabats of Table II, the thick lines the TFC adiabats, the thick dashed lines the TF adiabats, and the thin dashed lines are the interpolation adiabats of lead in accordance with^[7] and of aluminum in accordance with^[8].

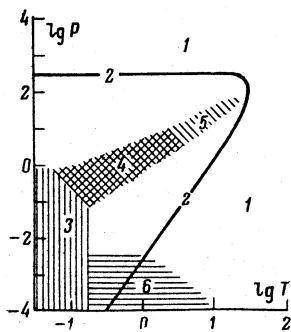


FIG. 4. Region of applicability of the TFC model—1, the limit of its applicability—2, region of static compressions—3, shock compression of solids—4 (absolute measurements) and 5 (comparative measurements), plasma experiments—6.

region of extremal states of the material. This makes it possible to construct, on the basis of the experimental data and TFC-model calculations, thermodynamically complete equations of state of any concrete substance.^[14]

¹⁾In the tables and in the figures, the pressure is in Mbar, the velocity in km/sec, and the temperature in eV.

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