Absolute measurements of shock compressibility of aluminum at pressures $P \gtrsim 1$ TPa

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(Submitted 19 December 1984)
Zh. Eksp. Teor. Fiz. **88**, 1452–1459 (April 1985)

A method was developed for the study of shock compressibility of dense media by simultaneously measuring the shock-wave velocity and the mass velocity of the material behind the front. The method is based on recording the instants when the collimating gap planes intersect with gamma-active reference layers implanted in the investigated substance. This method makes feasible, for the first time, sufficiently accurate "absolute" shock-wave investigations of dense-medium properties at "intermediate" pressures (0.5–15 TPa) and meets the need for deriving up-to-date equations of state. Specific experimental results were obtained for aluminum at pressures 0.93 and 0.99 TPa. The applicability of the method at 3.2 TPa is also verified. The results differ considerably from those obtained by the reflection method, so that corrections must be introduced into the interpolation equations of state.

I. INTRODUCTION

The shock compressibility of materials at $P \gtrsim 0.5$ TPa is measured mostly by the reflection method, in which the shock-wave front velocity is measured as the wave passes in succession through a standard material and the investigated one.[1–3] This method was used to investigate the shock compressibility of a number of materials in the range 0.5–400 TPa[4–11] At lower pressures, the equations of state of the standard substances are determined by absolute methods (e.g., by the deceleration method.[2] When substances such as aluminum are investigated at pressures $P \gtrsim 0.5$ TPa, one uses for the standard an interpolation equation of state, which is subject to a number of uncertainties. This makes the intepretation of the reflection-method data indeterminate.[7]

Absolute methods of determining shock compressibility are therefore particularly valuable. These are based, e.g., on simultaneous measurement of the shock-wave velocity D and of the velocity u of the matter behind the front. The mass, energy, and momentum conservation laws can yield directly the thermodynamic parameters of the matter on the shock adiabat, viz., the pressure $P = \rho_0 D u$, the compressibility $\delta = \rho/\rho_0 = D/(D-u)$, and the internal energy $\varepsilon = P(\delta - 1)/2\rho$ (ρ and ρ_0 are the density of the matter behind and ahead of the shock-wave front, respectively).

In Ref. 12 it was proposed to determine the mass velocity by measuring the Doppler shift of the resonance-capture lines of neutrons of energy 0.3–1.0 keV as they pass through a moving substance. The wave velocity was recorded using optical channels. Data were obtained on the shock compressibility of molybdenum at $P \approx 2$ TPa. In our opinion, however, the large measurement error ($\Delta D/D \sim \Delta u/u \sim 5\%$) prevents so far the use of this method to improve on the contemporary equations of state.

A method of simultaneously measuring the values of D and u with the aid of gamma-active reference layers was proposed in Ref. 13. The measurement error in this case is $\Delta D/D \sim \Delta u/u \sim 2\%$. We present here a substantiation of this method, discuss the feasibility of its use, present an experimental procedure, refine the earlier published data, and report new experimental results.

2. SUBSTANTIATION OF METHOD AND PHYSICAL SCHEME OF EXPERIMENTAL APPARATUS

Simultanous measurements of the wave and mass velocities on a shock-wave front is made possible by radioactive reference layers implanted in the investigated substance. The layers are parallel to the wave front. In the course of the gasdynamic motion, the reference layers are dragged by the moving substance. A system of collimating slits is used to determine the instant of their passage through control positions.

The simplest from the experimental viewpoint is a "plane geometry" of the shock-wave front, of the reference layers, and of the collimiating slits, wherein the corresponding surfaces are parallel to one another (Fig. 1). The required intensity and configuration of the employed shock-wave process can be produced by strong explosions. Measurements must be taken here to protect the collimating system from damage before the instants when the reference layer pass through the control points.

The experimental conditions must ensure, with high accuracy, planarity of the shock-wave front surface and of the region where the measurements are made. To this end, a cylindrical channel is placed in the path of the wave, made of a material (magnesium organic substance, and others) whose density is lower than that of the surrounding material. According to the results of two-dimensional gasdynamic calculations, the use of such a channel ensures that the inner shock-wave front will be far enough ahead of the front in the outer medium, and in the central part of the cylinder, whose diameter is $\sim 2/3$ of the outside diameter, the wave front will be plane. The measurement block is mounted on the end of the cylinder. Additional gasdynamic protection of the collimating system is provided by placing layers of dense material (lead, steel) in the path of the waves.

To measure the mass velocity u it suffices to regard the

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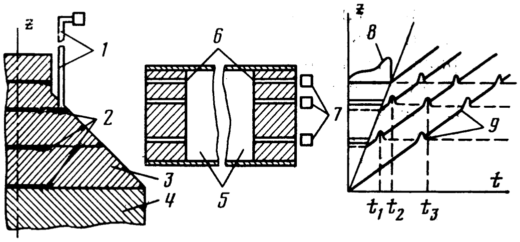

FIG. 1. Apparatus and plots of the recorded signals: 1—optic channel, 2—reference layers, 3—investigated substance, 4—shock-wave formation channel, 5—collimating system, 6—collimating slits, 7—gamma-ray detectors, 8—radiation signal of immobile reference, 9—radiation signals of moving references.

instants of passage of one reference layer through the planes of two collimating slits, the distance a between which is the measurement base. We have then $u = a/(t_3 - t_1)$ (see Fig. 1). Placement of two reference layers at two collimating slits makes it possible to determine D by comparing the starting instants of their motion: $D = a(t_2 - t_1)$. The shock compression δ on the wave front is determined then only from measurements of time intervals: $\delta = (t_3 - t_1)/(t_3 - t_2)$. The number of reference layers and collimating slits in experimental apparatus usually exceeds the indicated minimum. This yields information on the nonstationary behavior of the recorded shock-wave phenomenon and on the neutron and gamma processes that take place.

In the described and principally simple scheme, the basic element that makes temporal measurements possible is the reference layer. Its radiation must pass through peripheral layers of the investigated substance, in which the gasdynamic motion differs considerably from the employed "planar" shock-wave flow. The reference layer should therefore contain surces of hard gamma radiation or of fast neutrons ($E_n > 1$ MeV). Since the processes are fast and nonstationary, the layer radiation intensity must be ensure the possibility of recording the instants when they cross the planes of the collimating slit, using a detector such as a photomultiplier or a coaxial photocell in the analog regime.

It is impossible to use for these purposes stationary radiation sources. Even when a highly sensitive detector such as a photomultiplier (sensitivity $\alpha \sim 10^{-11}$ A $\cdot$ cm^2 $\cdot$ s/photon) is used, their activity must be $\sim 10^7$ Ci. Better suited for operation in strong backgrounds are coaxial photocells ($\alpha \sim 10^{-17}$ A $\cdot$ cm^2 $\cdot$ s/photon). The required source activity is then substantially higher. There are no such sources, but even if they existed, it would be impossible to work with their $\sim 10^4$ MW power requirement.

It is useful to consider pulsed sources that can exist during a certain stage of the employed gasdynamic process. In particular an intense γ-ray source can be obtained pulsed neutron irradiation of a substance whose nuclei have radiation-capture cross sections larger by $\sim 10^3$ times than the corresponding cross sections of the substance investigated. The existing pulsed sources usually produce fast neutrons ($E_n \sim 1$ MeV). The radiative-capture reactions proceed effectively at lower E_n. The neutron pulse must therefore precede the recorded gasdynamic motion by the time interval needed to moderate the neutrons in the investigated substance to the required energies. In a number of cases one can use in the reference layers europium, whose (n,γ) reaction cross section at $E_n = 10$–100 eV is $\sigma = 220$–80 b.

The standard substance widely used in the reflection method is aluminum. At "intermediate"pressures (0.5–15 TPa on the shock adiabat) the equation of state of this material exhibits substantial ambiguities due to the uncertainty in the allowance for the shell-electron effects in contemporary theoretical models.[14,15] Our experiments were therefore aimed at a study of the shock compressibility of aluminum.

Other reference + sample combinations are possible. In some cases pulsed neutron sources (converters can be used in the reference layers).

After the neutrons are moderated, the diffusion and absorption have pratically no effect on their density. We can therefore estimate the connection between the intensity Γ of the gamma reference [s^{-1}], on the one hand, and the fast-neutron flux Φ [cm^{-2}] and the mass M of the reference, on the other:

$$\Gamma \sim \Phi\sigma N v M/l \sim 0{,}2\cdot 10^7 \Phi M. \qquad (1)$$

Here σ is the cross section of the (n, γ) reaction on the reference-material nuclei, N is the number of these nuclei per unit mass of the reference, v is the velocity of the moderated neutrons, and l is the range of the external-flux neutrons in the investigated substance. The numerical value is given for aluminum and for Eu_2O_3, compressed to 2.7 g/cm^2, as the reference material. Since σ for europium decreases more rapidly than v^{-1} as the neutrons are moderated, the value of Γ will increase until it becomes equal to the rate at which the neutrons enter the reference layer from the aluminum and are absorbed by the europium. With further moderation of the neutrons, Γ will be decrease by the neutron depletion from the aluminum layers surrounding the reference layer. At a layer thickness ~ 0.5 cm the maximum intensity of its radiation is reached at $\sigma \sim 200$ b, corresponding to neutron energies $E_n \sim 10$–100 eV. The characteristic time of neutron moderation to these values of En is ~ 10–50 μs.

The neutron moderation in the sample heats the investigated material, and this influences its shock compressibility in the general case. For the example results to be easily interpreted it is necessary that this influence be weak. This imposes an upper bound on the flux Φ. Acceptable values for aluminum are $\Phi < 10^{17}$ cm^{-2}. The lower bound of the flux, as seen from (1), is imposed by the need to increase the size of the measurement block.

The reference layers act as an inhomogeneity in the investigated material, and are taken into account in the interpretation of the results on the basis of gasdynamic calculations. To decrease the uncertainties due to the introduction of calculation corrections it is necessary that the thickness of a reference layer not exceed 10% of the measurement base. The apparatus used in the experiment is capable of recording time intervals with acceptable accuracy ($\Delta t/t \sim 1\%$) at a duration ~ 2 μs. To study the shock compressibility of aluminum at a pressure ~ 1 TPa ($D \sim 25$ km/s) the base chosen was

~ 5 cm and the reference layer thickness $\Delta \lesssim 0.5$ cm.

The measurement block comprised a set of plates made of brand AD-1 aluminum (99% Al, $\rho_0 = 2.71$ g/cm^2), in which the reference layers were implanted in the form of wafers. This shape of the layer agrees with the cylindrical geometry of the shock-wave formation channel and makes it possible to place, when necessary, additional collimating systems. To ensure maximum recorded-radiation fluxes in the detector directions, the thickness of the investigated substance should be a minimum so as not to distort the front motion in the substance in the region behind the front where the references are located. The lateral surface of the block, on the side of the collimating system, was therefore made in the form of a cavity oriented at an angle 45° to the direction of the front motion. The front profile was monitored by three optical channels placed at the corners of an equilateral triangle on a 150-mm diameter (only one channel is shown in Fig. 1). We recorded the times of the light flashes upon emergence of the shock wave to a selected control surface perpendicular to the block axis.

The measurements were encumbered by an appreciable background. The main background source are the primary-beam neutrons, as well as the γ rays produced by (n, γ) reactions in the sample, in the structural elements of the experimental setup, and in the surrounding medium. Protection against neutrons is achieved by moving the detectors far from the investigated substance. Protection against captured γ radiation is provided by the collimating system, whose lead partitions attenuate completely the γ flux emerging from the measurement block to its environment. Only the radiation passing through the collimating slits remains unattenuated. If a coaxial photocell is used as a detector, a collimating-system length $L \sim 2$ m is sufficient. The detector current is connected with the intensity Γ of the radiation from the reference layer by the relation

$$I \approx \Gamma \alpha \exp(-\mu \rho d)/4\pi L^2,$$

where α is the sensitivity of the detector, μ the coefficient of mass attenuation of the γ rays by aluminum, ρ the density of the compressed aluminum, and d its thickness. To ensure $I \sim 1$ A at $\rho \sim 10$ g/cm^2, $d \sim 10$ cm, and $L \sim 2$ m a value $\Gamma \sim 10^{24}$ s^{-1} is necessary. In this case, according to (1), the mass of the part of the reference layer that radiates towards the detector should be ~ 50 g at a flux $\Phi \sim 10^{16}$ cm^{-2}. The reference material behind the shock-wave front is compressed to about one-third. The γ rays are then emitted effectively towards the detector from a reference-material depth 2–3 cm. We used in the experiments reference wafers of ~ 9 cm diameter and ~ 0.4 cm thickness.

The collimated system was an assembly of coaxial and rigidly interconnected lead disks in which 3×90 mm slits were cut. The slit width was governed by the characteristic thickness of the reference wafer compressed by the shock wave (~ 1 mm) and by the ease of aligning the collimating system. The distance between the slit planes are the measurement bases. The error in the determination of the sizes of these bases was governed by the manufacture imprecision as well as by the deviation from parallelism of the slit planes, the reference layer, and shock-wave front surface. The slit planes were mounted parallel to the reference-layer planes to within $\pm 0.2°$. The base error due to the deviation from parallelism is negligibly small compared with the imprecision of the system manufacture, which amounts to $\sim 0.2\%$ over a base of 50 mm. Monitoring the shock-wave front makes it possible, when necessary, to take into account the base-dimension correction necessitated by the nonparallelism of the front plane and the reference layers.

To lower the γ background recorded by the detectors prior to arrival of the shock wave, with an aim at determining D, the second and third reference layers (the reference layers and the collimating slits are numbered in the wave-propagation direction) were placed 1 mm deeper than the planes of the corresponding slits (see Fig. 1). The fourth reference layer was placed over the center of the slit, so that experimental information could be obtained on the course of the neutron and gamma-ray processes.

Placement of intense sources in the sample causes radiative heating of the substance. By the instant of arrival of the shock wave the interface between the aluminum and the reference is shifted and the states of these substances are altered. This influences noticably the interpretation of the experimental results, so that the corresponding processes must be taken into account by using gasdynamic calculations.

3. EXPERIMENTAL RESULTS

The experimental setups constructed in accord with the described principles were used to perform three experiments on the shock compresibility of aluminum. These setups differed in some detail (number of reference layers and of collimating slits, sizes of units and others).

The setup used in the first experiment had two collimating slits and two reference layers. The shock-wave profile was not monitored. The preliminary results of this experiment were reported in Ref. 13. The fact that these data deviated from the interpolation equations of state by amounts exceeding the measurement errors stimulated a search for the sources of the possible systematic errors not accounted-for in the primary data processing. The oscillogram-interpretation procedure was therefore improved by taking into account the peculiarities of the recorded signals, the distortions of the image, and others. Elimination of the systematic errors yielded new time intervals: $\Delta(t_2 - t_1)/(t_2 - t_1) \sim +4\%$, $\Delta(t_3 - t_2)/(t_3 - t_2) \sim -4\%$, $\Delta(t_3 - t_1)/(t_3 - t_1) \sim 0.9\%$. This led to more accurate experimental values of the shock-wave-front parameters averaged over a measurement base 5 cm: $\langle D \rangle = 24.8$ km/s, $\langle u \rangle = 15.2$ cm/s, and $\langle \delta \rangle = 2.57$. These values are the starting point of the subsequent data reduction.

Since the motion is nonstationary, the mass velocity of the material directly behind the shock wave differs from the reference velocity. The local parameters of the front at $D = \langle D \rangle$ were therefore determined by correcting the values of u and on the basis of gasdynamic calculations ($\Delta u/u \sim 2.3\%$, $\Delta\delta/\delta \sim 4\%$).

Substantial procedural refinements were also introduced, necessitated by allowance for the reference layers in

TABLE I.

Experiment No.	ρ_0, g/cm^3	D, km/s	μ, km/s	P, TPa	$\delta=\rho/\rho_0$
1	2,55±0,03	24,8±0,5	15,7±0,3	0,99±0,03	2,73±0,08
	2,71	24,2±0,7	15,1±0,4	0,99±0,03	2,65±0,1
2	2,58±0,03	24,0±0,4	15,1±0,2	0,93±0,02	2,70±0,05
	2,71	23,4±0,6	14,5±0,3	0,93±0,02	2,63±0,07
3	2,71	40±5	30±2	3,2±0,5	3,9±1,2

the measurement block. The intense γ radiation from this layers increases the internal energy of the aluminum to values $\varepsilon_0 \sim 1.5$ kJ/g. The ensuing pressure compresses the porous reference material to a density ~ 6 g/cm^3, which is accompanied in turn by a decrease of the aluminum density to ~ 2.55 g/cm^3. The corresponding corrections to u and δ were based on the calculations, and jointly with the corrections for the nonstationarity amounted to $\Delta u/u \sim 3.3\%$, $\Delta\delta/\delta \sim 6.2\%$. The final results are listed in Table I. The shock-wave-front parameters determined in this manner correspond to an initial-state aluminum shock adiabat at an initial state with $\rho_0 = 2.55$ g/cm^3 and $\varepsilon_0 = 1.5$ kJ/g.

To check on the correctness of the indicated procedural refinements, a second experiment was performed at the same pressures as in the first, but with a more extensive test program. The experimental setup is shown in Fig. 1. In particular, the third collimating slit verified the ideas concerning the neutron and γ processes and the behavior of the reference layers. An oscillogram from a detector placed in this slit is shown in Fig. 2. The pulse produced by the primary neutrons is followed by a growing γ-ray signal from a fourth reference layer. The decrease of the growth rate in the t interval ~ 13–21 μs is due to compression of the reference layer by the expanding aluminum, which leads in turn to an increase of the range of the γ rays in the reference material. The change of the character of the growth of $\Gamma(t)$ at $t \sim 26$ μs (inflection point) is due to the start of the equalization of the rates of entry of the neutrons into the reference layer and of their absorption by europium. Since the fourth reference layer was intended for procedural investigations, it received the partly relaxed shock-wave front. The decrease of $\Gamma(t)$ at $t > 31$ μs is therefore stretched-out somewhat and consists of two sections of durations ~ 2 and ~ 0.5 μs with different fall-off rates. The reference leaves the slit boundary at the instant $t \sim 33$ μs. The detector records subsequently the passage of deeper reference layer against a growing radiation background. The reference intensities estimated with allowance for the real experimental conditions amount to $\Gamma_1 \sim 5.10^{24}$ s^{-1}, $\Gamma_2 \sim 10^{24}$ s^{-1}, $\Gamma_3 \sim 0.4.10^{24}$ s^{-1}, $\Gamma_4 \sim 0.5.10^{23}$ s^{-1}.

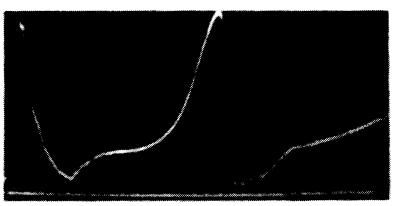

FIG. 2. Oscillogram of signal from detector placed in the third slit. The time between markers is 2 μs.

The oscillograms, with a short time scan, are similar to that shown in Ref. 13. We measured in the experiment the following time intervals: $t_2 - t_1 = 2.08 \pm 0.04$ μs, $t_3 - t_2 = 1.31 \pm 0.02$ μs, $t_3 - t_1 = 3.39 \pm 0.05$ μs. The errors correspond to a probability confidence level 0.95. The time intervals between the remaining pulses, which characterize the nonstationary character of the shock-wave process, agree with the theoretical-calculation assumptions concerning the investigated phenomenon.

The data obtained with the optical channels have confirmed that the deviations of the shock-wave front from a plane is negligible. The oscillograms are similar in character and quality to the one shown in Ref. 11. The maximum difference in the time required for the wave to reach the control plane was 0.02 μs. The experiment yielded the values $\langle D \rangle = 24.0$ km/s, $\langle u \rangle = 14.75$ km/s and $\langle \delta \rangle = 2.59$.

The additional data reduction needed to allow for the nonstationary character of the wave and of the inhomogeneity of the medium, was similar to that in the first experiment. The corresponding summary corrections were $\Delta u/u \sim 2.4\%$ and $\Delta\delta/\delta \sim 4.2\%$. The decrease of the corrections compared with the first experiment is due to the smaller effect of the deviation of the wave from stationary. The results of the final reduction are listed in Table I. These data correspond to an aluminum shock adiabat from an initial state with $\rho_0 = 2.58$ g/cm^3 and $\varepsilon_0 = 1.5$ kJ/g. The deviation of ρ_0 from the first experiment is due to the smaller thickness of the reference layer.

The third experiment was aimed at studying the shock compressibility of aluminum at higher pressures ($P \sim 3$ TPa). The feasibility of such experiments was confirmed by the experiment, but only order-of-magnitude oscillograms were obtained. The estimated time intervals for an 8-cm base are: $t_2 - t_1 = 2.0 \pm 0.2$ μs, $t_3 - t_1 = 2.7 \pm 0.2$ μs, $t_3 - t_2 = 0.7 \pm 0.2$ μs. The shock-wave front parameters obtained from these data are given in Table I.

4. DISCUSSION AND RESULTS

In numerous experimental compressibility studies the aluminum and an initial density 2.71 g/cm^3. The interpolation equations of state were therefore deduced and compared with one another by invoking shock adiabats corresponding to this density. It was of interest to compare our results with these data. This can be done by by calculating the start from different initial states. In particular, in the cases investigated the correction to compression at constant pressure is small and amounts to $\Delta\delta/\delta \approx 3\%$. Its value dependslittle on the

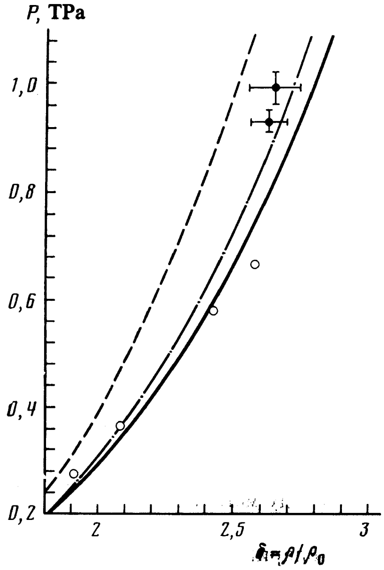

FIG. 3. Shock adiabats of aluminum: solid curve—from the equation of state of Ref. 17; dash-dot—interpolation relation from Ref. 7; dashed—results of calculations of Ref. 16; light circles—experimental results of Ref. 7; dark circles—our results.

type of the equation of state, and the compression scatter is in this case $\sim 1\%$. By using such a recalculation it is possible to represent the obtained experimental information in the form of points on the shock adiabat corresponding to an initial density 2.71 g/cm^3. The errors in the determined front parameters are increased by the uncertainties of the recalculation. The corresponding data are given in Table I and in Fig. 3.

For comparison with the results obtained by other, Fig. 3 shows aluminum shock-adiabat points obtained by the reflection method, with quartzite as the reference.[7] The figure shows also the interpolation shock adiabat proposed in the same reference, the theoretical shock adiabat obtained in Ref. 16, and also the shock adiabat constructed with the aid of the interpolation equation of state in Ref. 17. It can be seen that both experimental curves obtained in the present paper are in good agreement and are somewhat on the "harder" side of the interpolations of Refs. 7 and 17, but on the "softer" side of the shock adiabat of Ref. 16. The agreement with the shock adiabat of Ref. 7 can be regarded as satisfactory, but the interpolation equation of Ref. 17 needs correction. A substantial difference between our results and the data of the reflection method[7] is observed. Since aluminum is a valuable material for shock-compression investigations, a critical review of the reflection-method data in the pressure region in question is necessary, as is also in experimental chack on these data.

In light of the results it would be of great value to use the proposed method to measure the shock compressibility of aluminum both at pressures ~ 0.5 TPa to refine the results of the reflection method, and at 3–15 TPa to study the influence of shell effects.[14,15] Unfortunately in view of the large errors, the results of the third experiment cannot be used for this purpose.

The described experiments are the first applications of the reference method of measuring shock compressibility. The experience with this method shows that there are great prospects of improving it both by using different combination of references and investigated materials and by differntly organizing the experiments.

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Translated by J. G. Adashko