Peculiarities of the phase transformation of bismuth in a rarefaction wave

M. N. Pavlovskiĭ and V. V. Komissarov

(Submitted 5 January 1982; resubmitted 2 August 1982)
Zh. Eksp. Teor. Fiz. **83**, 2146–2151 (December 1982)

Results are presented of experimental recording of a two-wave configuration of a shock front in bismuth, which attests to occurrence of a first-order phase transition in bismuth at a pressure 2.55 GPa. A shock transition is observed in a rarefaction wave propagating in bismuth shock-compressed to 5.8 and 9.28 GPa. The amplitude of the rarefaction shock wave is 1.9 GPa. It is shown thus that an inverse phase transition takes place in a rarefaction wave at pressures noticeably lower than the critical polymorphic-transformation pressure in a compression wave.

PACS numbers: 62.50. + p, 64.70.Kb

It was shown in Ref. 1 that when a first-order phase transition is produced in the course of relaxation of shock-compressed ionic KCl and KBr crystals the amplitude of the rarefaction shock wave is much lower than the critical pressure of the phase transformation of these substances in a compression wave. It seemed expedient to study this phenomenon in solids of other classes, for example in bismuth. It is known that under static compression bismuth undergoes a first-order phase transition at a pressure 2.58 GPa.[2] If a two-wave shock-front profile were recorded in bismuth, this would be evidence of a polymorphic transformation in it also under dynamic loading conditions. The profile of the loading wave and later also the profile of the rarefaction wave in bismuth was recorded by us with a manganin pressure sensor by the procedure described in Ref. 3.

Manganin sensors in the form of bifilar coils of ~ 4.5 mm diameter were secured with epoxy resin between two plates of chemically pure bismuth (see Fig. 1) whose density ρ_0 was 9.80 g/cm^3. The sensors were insulated from the bismuth by mica plates ~ 0.02 mm thick. The thickness of the manganin sensors was ~ 0.03 mm. The pulses feeding the sensors were turned on ~ 10 μsec before the arrival of the loading shock wave. The recorded sensor-voltage pulse was fed directly to the deflecting system of an S9-4 oscilloscope. To be able to disregard the change of the resistance of the manganin-sensor leads during the compression, the voltage was applied to it with a special pair of leads. The recorded signal was picked off the sensor with an identical pair of leads. To make fuller use of the working part of the oscilloscope screen and to increase the measurement accuracy, a rectangular voltage pulse of approximate amplitude 10 V from a G5-15 generator was applied to the deflecting system of the oscilloscope and shifted the beam to the null line (see the oscillograms in Figs. 2–4). The frequency of the timing sinusoid on the oscillograms, 10 MHz, was monitored with a Ch3-34 frequency meter. The asterisks on the oscillograms mark the instants when the shock was closed the contacts. To reveal the configuration of a compression these contacts were placed (see Fig. 2) on the boundary between the screen and the bismuth sample, and to record the profile of a rarefaction waves they were placed on the free surface of the sample.

The bismuth shock-compression pressure P was determined from the experimentally measured electric resistance of the manganin sensor in the compressed state, calculated from the formula

$$R=R_0(Z_0-Z_1+Z_2)Z_0^{-1},$$

where R_0 is the initial resistance of the manganin pickup, equal to $\sim 1.5\Omega$; Z_0, Z_1, and Z_2 are the amplitudes of the oscilloscope-beam deflections. To convert from R/R_0 to P we used a pressure dependence of the manganin resistance[3] close to those given in Refs. 4 and 5:

$$P=32.41\frac{R-R_0}{R_0}+12.35\left(\frac{R-R_0}{R_0}\right)^2-5.69\left(\frac{R-R_0}{R_0}\right)^3\text{GPa}.$$

The experimental results have shown that the amplitude of the first compression wave, equal to the pressure of the phase transition in bismuth, amounted to 2.55 ± 0.05 GPa (wave velocity $D_1 = 2.03$ km/sec, mass velocity $U_1 = 0.135$ km/sec, density of material $\rho_1 = 10.5$ g/cm^3 behind the shock-wave front); this is very close to the value obtained in Ref. 6.

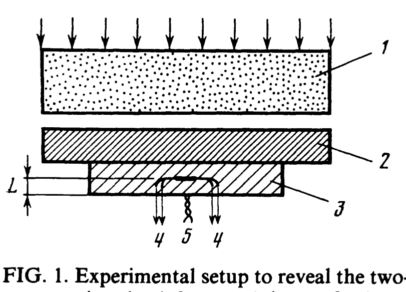

FIG. 1. Experimental setup to reveal the two-wave configuration of the compression shock front and the rarefaction wave profile in bismuth: 1—explosive charge of 90 mm diameter, 2—metallic screen 10 mm thick, 3—bismuth sample, 4—manganin pressure sensor, 5—electric contact-making sensor.

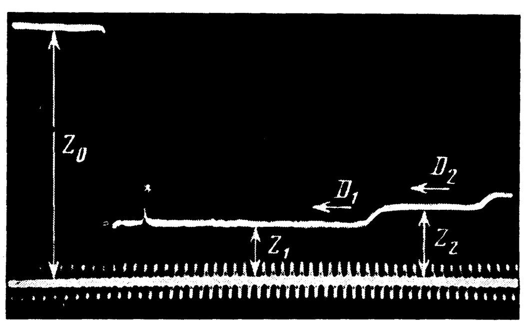

FIG. 2. Two-wave configuration of shock front in bismuth. Amplitude of first compression wave 2.55 ± 0.05 GPa, of second wave 4.4 ± 0.1 GPa.

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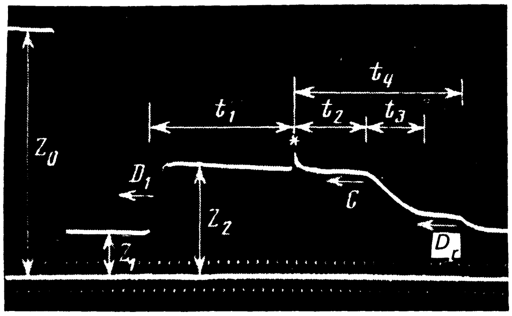

FIG. 3. Profiles of compression and rarefaction waves in bismuth. D_1—compression shock wave of amplitude 9.28 GPa, C and D_r—elastic-plastic and rarefaction shock waves.

The ultrafast low-temperature recrystallization recorded in this case allows us to assume that under shock-compression conditions the predominant feature is the martensitic character of the transformations, known[7] to be based on the collective displacement of many atoms through short distances along the cleavage planes. Such a displacement requires no thermal activation and is completed within times on the order of 10 nsec. Since the mechanism of formation of dense phases in static and shock compressions of solids can be different in principle, one should expect also differences in the critical pressures of the phase transitions under different compression conditions. The equality of the pressure of the start of the phase transition in the static case to the experimentally measured uniaxial stress in the dynamic case is evidence that the bismuth is in a state close to hydrostatic behind the shock-wave front.

The experimental setup for the study of the rarefaction-wave structure in bismuth was similar to that shown in Fig. 1. The amplitude of the shock-compression wave was in this case 9.28 GPa. The initial state of the bismuth in the relaxation experiments correspond thus to two polymorphic transformations—at 2.58 and 7 GPa,[8] and possibly also at 4.27 GPa (Refs. 9, 10). The wave velocity D_1 in bismuth, namely 2.12 km/sec, was determined by starting from the sample thickness $L = 2$–5 mm and the measured time interval t_1 between the instant of arrival of the compression shock wave at the manganin sensor and the instant of closing of the contact-making sensor mounted on the free surface of the bismuth sample. The mass velocity of the matter behind the compression shock wave was $U_1 = 0.45$ km/sec.

Knowledge of the degree $\rho_1/\rho_0 = 1.267$ of relative compression of the material by the compression shock wave, of the thickness L, and of the time interval t_2 between the instant when the shock wave closed the contact-making sensor and the instant of arrival of the rarefaction wave at the manganin pickup made it possible to determine the maximum propagation velocity C_1 of the rarefaction shock wave through the shock-compressed bismuth:

$$C_1 = \frac{L\rho_0}{\rho_1 t_2} = 2.9 \text{ km/sec}.$$

From the time interval $t_2 + t_3$ (see Fig. 3) we determined, with the aid of the constructions on the x-t diagram (see Fig. 5), the propagation velocity of the tail part of the plastic rarefaction wave, $C_2 = 2.03$ km/sec. The mass velocity $U_2 = 0.83$ km/sec behind the plastic rarefaction wave was obtained by extrapolating, on the P-U diagram, the mirror image of the shock adiabat of the dense phase of bismuth from the point with $P_1 = 0.28$ GPa to the pressure behind the plastic rarefaction wave. As is well seen from the oscillogram of Fig. 3, the rarefaction wave in bismuth contains a clearly pronounced shock transition with an amplitude ~ 1.9 GPa. This quantity includes a correction[11] for the hysteresis[12] of the manganin ($\sim 1.6\%$) in the relaxation wave.

It is impossible to explain the abrupt pressure jump observed by us (see Figs. 3 and 4) in the rarefaction wave as being due to solidification of the bismuth after its melting in the compression shock wave, inasmuch as at a pressure 9.28 GPa the temperature is insufficient to melt the shock-compressed bismuth. This is indicated also by a comparison of our results with measurements[13] of the speed of sound behind the shock-wave front in bismuth. It is known from Ref. 13 that melting of bismuth in a shock wave called for heating of the bismuth sample to a temperature 473 K prior to the shock compression.

The study of bismuth has thus shown, just as earlier in

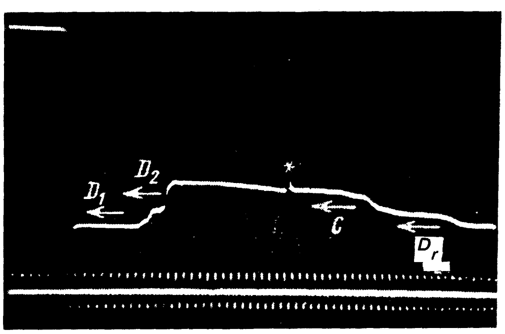

FIG. 4. Profiles of compression and rarefaction waves in bismuth: D_1—compression shock wave of amplitude 2.5 GPa, D_2—second compression shock wave of amplitude 5.8 GPa, C and D_r—rarefaction elastoplastic and shock waves.

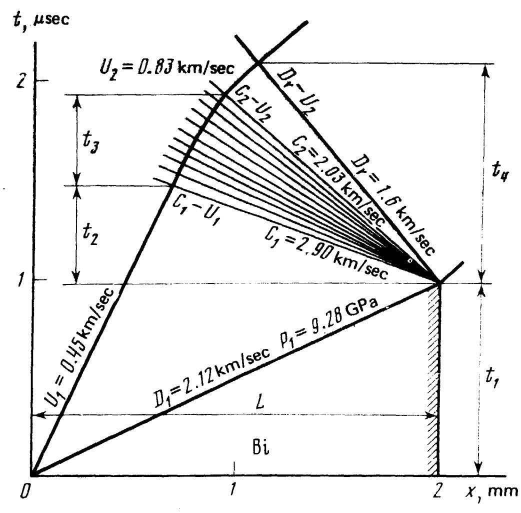

FIG. 5. x—t diagram of the processes of shock compression and subsequent relaxation of bismuth.

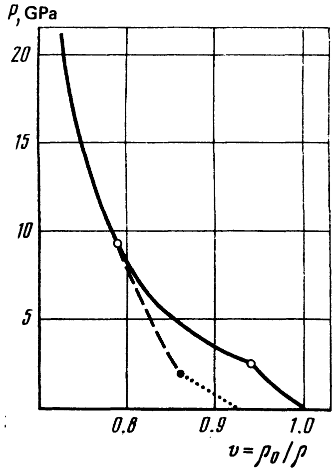

FIG. 6. $P(v)$ plot of bismuth: solid curve—experimental shock adiabat of bismuth from Ref. 15, dashed curve—calculated expansion isentrope, dotted curve—rarefaction shock wave ($D_r = 1.6$ km/sec).

the case of KCl and KBr, that the inverse phase transition in the relaxation wave takes place at pressures noticeably lower than the critical phase-transition pressure in compression. Recording the time interval t_4 from the instant when the contact-making sensor closed to the instant of arrival of the rarefaction shock wave at the manganin sensor made it possible, after drawing the required plots on the x-t diagram (Fig. 5), to determine the rarefaction shock-wave propagation velocity in bismuth, $D_r = 1.6$ km/sec. To verify that the rarefaction shock wave observed by us is not connected with the phase transition that occurs at ~ 7 GPa (Refs. 8 and 9), an additional set of experiments was performed, in which the amplitude of the shock compression of the busmuth was ~ 5.8 GPa. As seen from oscillogram of Fig. 4, the compression front has in this case a two-wave structure. The amplitude of the first compression wave is ~ 2.6 GPa. The rarefaction wave contains as before the shock transition. Its amplitude and propagation velocity are close to those measured earlier at $P_1 = 9.28$ GPa.

Starting with the obtained valued of P_1, C_1, C_2, U_1, and U_2 we estimated the positions of the bismuth expansion isentrope from the point of the shock adiabat with pressure $P_2 = 9.28$ GPa. The calculation was based on the fact that the following relations hold in the rarefaction wave[14]:

$$\Delta P = C^2 \Delta\rho, \quad \Delta U = \frac{C\Delta\rho}{\rho}.$$

Specifying a certain value of $\Delta\rho$, we calculate the corresponding values of ΔP and ΔU. Next, knowing ΔU and using the relation $C = f(U)$ obtained by us, we find the new value C' of the sound velocity. We obtain thus the parameters $P_1 - \Delta P$ and $\rho_1 - \Delta\rho$ of the first point of the expansion isentrope. Specifying the next value of $\Delta\rho$ and performing the described calculation in the same sequence as before, we determine the parameters of the next isentrope point. Advancing successively from point to point we reveal the course of the bismuth expansion curve all the way to the shock-transition pressure. The position of the isentrope is shown in Fig. 6.

The wave ray corresponding to a rarefaction shock wave with $D_r = 1.6$ km/sec and drawn from the isentrope point with pressure 1.9 GPa crosses the volume axis on Fig. 6 far to the left of the point v_0. These experimental data indicate that the bismuth density behind the rarefaction shock-wave front is substantially higher than the initial density ρ_0. This allows us to assume that not all the bismuth manages to undergo in the rarefaction shock wave the reverse transition to the initial phase. It is also possible that the transition in the rarefaction shock wave is not to the initial phase but to some intermediate phase which then goes over more slowly to the initial one. This is attested by the results of an x-ray structure analysis of bismuth samples preserved by the procedure described in Ref. 16 after shock compression to pressures ~ 12 GPa; these have shown the presence of only one initial structure of bismuth.

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