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RELAXATION EFFECTS IN SHOCK-INDUCED TRANSITIONS IN BISMUTH

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INTRODUCTION

The transition from Sb I to the II-III mixed-phase region is accompanied by a large volume change that can appear under shock compression as a double wave structure[1,2]. This transition occurs sluggishly on a time scale of a few microseconds. The I-II transition in Bi, its semimetal neighbor in the Group V elements, is accompanied by a large volume change that occurs on a nanosecond or subnanosecond time scale under similar conditions[3]. Although the structures for phase I for both elements are the same--namely the rhombohedral (2 atoms/unit cell) As type--those in phase II are different[4,5]. Some investigators have suggested that the structures in phase III, however, are again alike or at least very similar[4,5,6]. If this is true, the question of whether Bi exhibits similar kinetics in the I-III transition arises. The higher pressure Hugoniot for Bi is shown in Fig. 1. Results for pressures greater than 42 GPa were not included because of a phase change postulated to occur at that pressure[7]. The solid line through the data is a shock velocity-particle velocity fit of the form $U = 1.140$ km/s + 2.134 u for an average density of 9.81 Mg/m^3. The limiting Rayleigh line, which corresponds to a shock velocity of 2.089 km/s, passes through a I-II shock transition stress of 2.54 GPa and a volume compression ratio of 0.9405 --an average of the values obtained by Larson using quartz gauges and Asay using a velocity interferometer[3,8]. An expanded picture of the mixed region is shown in Fig. 2. The Rayleigh intercept of the high pressure Hugoniot and the second shock data of Hughes et al.[9] and of Duff and Minshall[10] were used to describe a Hugoniot fit through the mixed region of the form $U = 1.00$ km/s + 1.43 u + 4.92 u^2 s/km. This fit applies in the stress region from 2.54 GPa to 9.1 GPa and is based on an average initial density of 10.40 Mg/m^3, the density at the I-II transition point. For each experiment included, the stress and compression for the second shock were determined with respect to the variables actually measured for the leading shock in that particular experiment. The data are presented, however, with respect to the precise measurements of Larson and Asay. This approach of using measured differences was adopted because of problems in determining the absolute velocity of leading waves in the earlier experiments[11]. Included in the figure for comparison is the room temperature isotherm measured by Giardini and Samara[12]. The phase notation is that of Klement et al.[13]. The [illegible] angular points, which represent recent data by Romain[14], are the only other previous results in this regime. Romain claimed to have observed a double wave structure between 7.0

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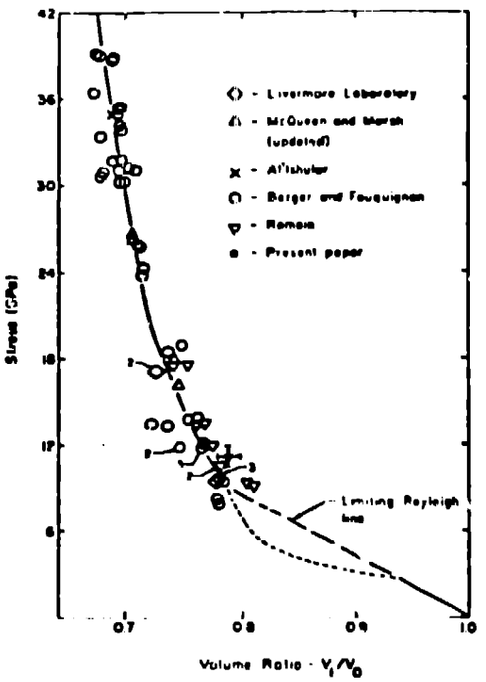

Fig. 1

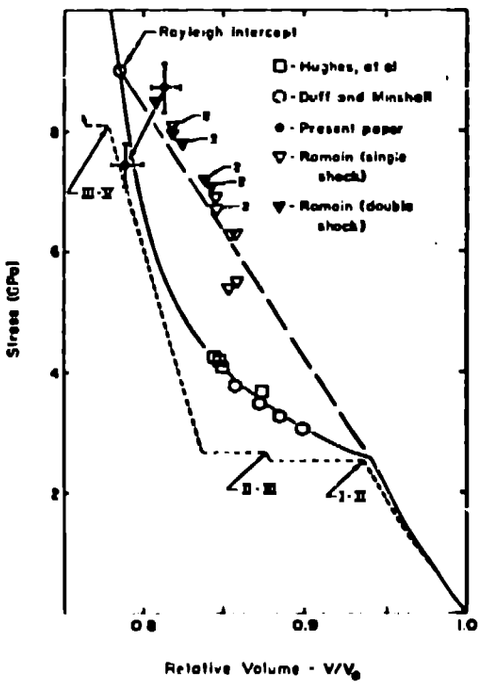

Fig. 2

and 8.6 GPa that could be explained by a shock-induced phase change at 7.0 ± 0.5 GPa. He postulated this to be the III-V transition. His data, on samples typically 1-2 mm thick, were obtained by observing electrical signals caused when a shock crossed a bimetallic junction--a technique not yet in widespread use. Two results in Fig. 2 lie below the precisely determined Rayleigh limiting line. In the one case where a specific experiment appears to have been duplicated with sample thicknesses of nominally 1, 2, and 3 mm, the second shock appears to decay in velocity 2.5% per mm as it traverses the sample. Romain, in his analysis, assumed no such behavior. Thus, there are numerous reasons why the Bi II-III mixed-phase region deserves more study.

RADIOGRAPHIC EXPERIMENTS

A sample compressed obliquely by a detonation sweeping along the sample-explosive interface can, under certain circumstances, show a double wave structure discernible by flash radiography. Details of this technique are described elsewhere[2,15,16]. The line tracing of a radiograph of a Bi sample driven by TNT presented in Fig. 3 is illustrative of this method. When the detonation has been permitted to travel sufficiently far, the wave structure becomes independent of time--at least within the resolution of the measurements. The radiograph then provides a kind of time history of the wave evolvement. For a double shock structure, the shock and particle velocities are

related to the stress and volume before (i-1) and behind (i) a shock by

$$U_i = V_{i-1}\sqrt{(\sigma_i-\sigma_{i-1})/(V_{i-1}-V_i)} \quad \text{and} \quad u_i = \sqrt{(\sigma_i-\sigma_{i-1})(V_{i-1}-V_i)}$$

When the detonation velocity is known and steady, these velocities may be obtained directly from the radiograph by measuring the angles indicated. Details of this technique are described elsewhere[16].

The Bi used in these radiographic studies was a cast material with a density of 9.77 Mg/m^3. Flash radiographs were taken at the PHERMEX facility[17]. Elastic precursor effects were ignored, both because they would not alter the results within the significance of the measurements and also because they could not be detected radiographically. In the first experiment TNT was used as the driving explosive. A line tracing of the radiograph is on the left side of Fig. 3. Both interface and intersecting shock exhibit gentle continuous curvature as a result of the decay caused by the release wave that follows the detonation. The result for the shock at the interface [$\theta = 19.88 \pm 0.43°$ and $\delta = 3.95 \pm 0.22°$] is shown in Fig. 1. The pressure at the interface is sufficiently high that no double shock structure occurs initially. After the pressure decays and the double structure forms, the leading shock is straight because the rarefaction follows the second shock. The velocity of the leading shock is 2.083 ± 0.009 km/s [$\theta = 17.57 \pm 0.08°$], which is within the standard deviation of that expected. No other shock structure is observed during the decay of pressure through the regime investigated by Romain. The second experiment used a baratol driver to investigate a Bi sample with a minute crack parallel to the sample-explosive interface. The bend in this crack was used to directly measure the compression ratio across the lead shock. The measured bend of $1.44 \pm 0.41°$ indicated a volume ratio of 0.937 ± 0.018, which agreed with the previous more precise work and confirmed the lead shock to be as expected. In a subsequent experiment the known velocity of this shock was then used to calibrate the detonation velocity along the interface, an approach analogous to the way that the I-II transition pressure is used to statically calibrate a pressure scale. The shock structure near the interface is shown in the line tracing in the right side of Fig. 3. The junction of the lead shock and second shock is not at the interface, but is connected to it by a short stem section that corresponds to a shock run of about 2 mm. A measurement of the angles associated with the stem lead to a stress prediction of 8.7 GPa [$\theta = 27.67 \pm 0.20°$ and $\delta = 4.57 \pm 0.23°$]. This is the higher pressure result shown in Fig. 2. In the double structure the velocity of the second shock can be measured directly. In Fig. 2 the resulting state would lie below the limiting Rayleigh line. Because no bend is discernible in the interface as a result of the transition from stem to double structure, the deflection of the interface can be used to characterize this state directly. With this approximation, the stress behind that shock is

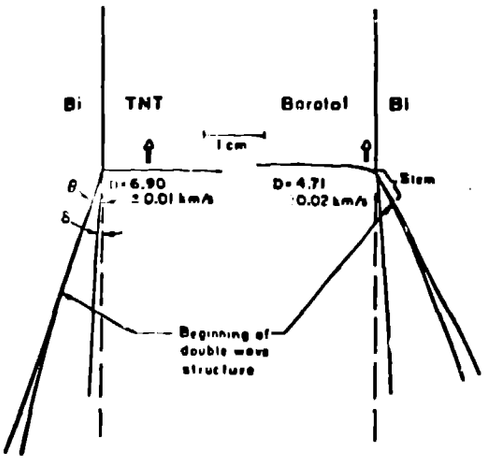

Fig. 3

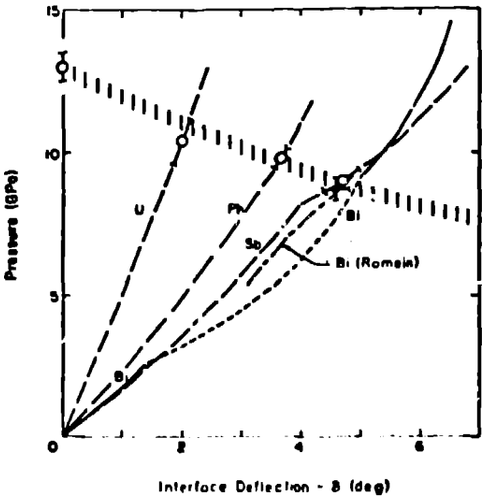

Fig. 4

determined to be 7.4 GPa [θ = 23.00 ± 0.34°]. This state is also shown in Fig. 2. The result is that the shock initially lies in the regime of Romain's data but, as it travels further into the sample, decays in 0.9 μs along the direction of the arrow to the mixed-phase Hugoniot. Because all but one of Romain's samples were thinner than 2.3 mm, these results are not necessarily in disagreement.

The reasonableness of this pressure interpretation has been checked by a somewhat independent method. When a sample is driven obliquely, the deflection of the explosive-sample interface, across which the pressure must remain continuous, is accompanied by an expansion of the detonation products. Thus the shock strength in a sample is directly related to that deflection. This phenomenon has been detailed elsewhere for other explosives[16]. Results for baratol are shown in Fig. 4. Zero deflection corresponds to the CJ pressure. An experimental observation of the shock in U [θ = 36.2 ± 0.3°], coupled with low-pressure Hugoniot data[18], and of the shock in Pb [θ = 32.48 ± 0.17°], interpreted with a previously used Hugoniot[16], added two other peg points. The Sb result, which came by extrapolating a double shock structure to remove relaxation effects, was included because of its proximity to the Bi result, but is too complex to be described here. The band of short vertical lines defines baratol behavior. The difference between the standard Bi mixed-phase Hugoniot, which is shown as a dotted curve, and that of Romain's data is apparent. The measurement of the interface deflection alone places the stem result with Romain's results.

It is important to not overlook the fact that baratol has a finite reaction-zone length that might affect the dynamic flow. No such effect, however, was noted in Pb, a standard well-understood material, so the kinetic feedback would have to be quite complicated

to influence Bi in a pressure regime only slightly different.

CONCLUSION AND RECOMMENDATION

The indication given by the baratol experiments is that as the shock in the mixed region progresses and decays, the lead state does not change along a stable Hugoniot but appears to change from a sort of metastable Hugoniot, in accordance with Romain's data, to a stable Hugoniot reminiscent of the earlier results. If this is so, then the Bi I-III transition is exhibiting a relaxation effect similar to that seen in the Sb I-III transition. The transition detected by Romain could then be interpreted as the II-III transition seen in the metastable region. The few experiments outlined here merely hint at an explanation. Additional precision work in the stress range 6-10 GPa, some with thin samples, is needed to resolve this behavior.

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