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MEASUREMENT OF AN EQUATION-OF-STATE POINT FOR MOLYBDENUM AT VERY HIGH PRESSURE*

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ABSTRACT

The Los Alamos Scientific Laboratory has conducted a variety of scientific experiments associated with nuclear explosions. Nuclear physics experiments have been especially well exploited in the past and have been reported at an earlier PNE meeting. A recent scientific application of nuclear explosions involved the development of a method to measure equation-of-state properties of materials behind strong shocks. A shock was generated by rapidly fission heating a slab of ^{235}U by exposing it to neutrons at $\sim$ 1 m from a nuclear explosion and allowing it to expand into an adjacent sample of the material to be studied. A point on the molybdenum equation of state was determined at a pressure of 2.0 TPa (20 Mbar) by measuring directly both the shock velocity and the particle velocity behind the shock. The shock velocity was obtained by measuring the transit time of the planar shock between positions at two depths in the molybdenum. This time interval was determined by observing the light flashes produced when the shock reached free surfaces located at the two depths. The particle velocity was obtained by observing the Doppler shifts of six neutron resonances in the neutron energy region from 200 to 800 eV in the moving,

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shocked molybdenum. The pressure and density derived from this pair of velocity measurements, made to an accuracy of $\pm$ 5%, are 20 Mbar and 25 $g\text{-}cm^{-3}$, respectively. This experiment represents the first direct determination of a point on the equation-of-state of any material in this pressure region, and the resulting data point is in good agreement with theoretical estimates. It appears that errors in both the shock velocity and the particle velocity can be reduced to approximately $\pm$ 2% in an improved measurement, resulting in a well-defined point on the equation of state for molybdenum, which can be used as a standard in future impedance-matching experiments.

I. INTRODUCTION

At the Los Alamos Scientific Laboratory we have been interested in scientific applications of nuclear explosions for many years. There have been a variety of scientific applications, among which have been meteorology studies involving tracking of airborne radioactivity, seismology, nuclear chemistry including heavy element production, nuclear physics, and shock phenomena. The United States has carried out several experiments involving heavy element production[1-8] with nuclear explosives, and the elements einsteinium and fermium were discovered in this way.[9] The Los Alamos Scientific Laboratory has conducted many nuclear physics experiments that have been extensively reported,[10] some of which were discussed at the Third Peaceful Nuclear Explosions Meeting.[11] The study of physical properties of materials at the high pressures such as those generated by nuclear explosives has been reported by workers in the U.S.S.R.[12-16] Besides furthering our understanding of the behavior of materials under extreme conditions, experiments at high temperatures and pressures are pertinent to problems of astrophysical and geophysical interest.

As part of the Los Alamos program of scientific applications of nuclear explosions, a new technique has been developed to study high-pressure shock phenomena and this technique will be the subject of this paper.

II. EXPERIMENTAL METHOD

Under ideal shock conditions, in planar geometry, all material ahead of the shock is stationary and at low temperature and pressure. Behind the shock front the material has been heated by the shock and is moving with a uniform velocity (particle velocity) that is less than the velocity of the shock front (shock velocity). The material behind the shock is at a constant high pressure and temperature and has been compressed to a higher density than the undisturbed material ahead of the shock. Determination of the pressure, density, and internal energy behind the shock constitutes a measurement of one point of the equation of state of the material. In the pressure region of interest here (tens of millions of atmospheres) ordinary solids are substantially compressed and heated. In this experiment, a sample of molybdenum was compressed to 2.5 times normal density by a pressure of twenty million atmospheres and simultaneously heated to a temperature of $\sim$ 70,000 K.

It can be shown by the use of the conservation relations for matter, momentum, and energy that the quantities pressure, density, and internal energy behind the shock are determined if both the hock and particle velocities can be measured. Measurement of the shock velocity is straightforward since it involves only a measurement of the time interval required for the shock to pass through different thicknesses of the material. At the high temperature of the shocked material in this experiment, the metal is incandescent; the flashes of light produced as the shock emerges from free surfaces at different depths in the sample are easily detected and used to measure the transit time and hence the shock velocity.

The problem of measuring the particle velocity is much more difficult. At relatively low pressures, several indirect methods are used but these are not practical in our experiments. In the Soviet experiments, a high-pressure shock was transmitted through a standard material into the material to be studied. If one assumes that the properties of the standard are known, then in this impedance-matching technique measurements of the two shock velocities in the two materials suffice to determine the properties of the unknown material. At the pressures in the experiments conducted by workers in the U.S.S.R. shock properties had not been experimentally determined for any material, and they relied upon theoretical extrapolation of low-pressure data for determination of their standards.

In order to measure the particle velocity it is necessary to probe into the matter behind the shock. In principle, either gamma rays or neutrons could be used. We chose to use neutrons as a probe because of the convenient characteristic of neutron resonances -- namely, for most materials neutrons are strongly absorbed at certain resonant velocities. On the other hand, neutrons with other velocities interact only weakly, thereby furnishing a penetrating probe. The resonant velocity, however, refers to the velocity of the neutron relative to the absorber nucleus. In some materials, resonances occur at neutron velocities comparable to the particle velocity that we wish to measure. Thus, if the absorber is moving in the same direction as the neutron, the resonance will be Doppler shifted and will occur at a higher neutron velocity than for a stationary absorber. In particular, if a shock has penetrated part way through a sample of material with sharply defined resonances and a burst of neutrons of all energies is sent through the sample, then the moving part of the sample will absorb neutrons at sharply defined velocities while the stationary material will absorb neutrons of lower velocities. The difference between the velocities, for corresponding resonances, is just the particle velocity that we want to measure.

It remains only to measure the velocity spectrum of the transmitted neutrons to locate the dips in the spectrum corresponding to the absorbing resonances. Since part of the sample is moving and part is at rest, each resonance occurs at two velocities separated by the particle velocity.

We chose to study molybdenum for three reasons: It has absorption resonances at convenient neutron velocities; it has a high boiling point so that it can withstand the considerable heating caused by neutrons passing through it prior to shock arrival; and its equation of state is well known at lower pressures so that we can predict its behavior at higher pressures with some confidence. In order to generate a shock in molybdenum, a disk of ^{235}U, 11-mm thick and 178 mm in diameter, was placed one meter from a nuclear explosive. A 2.5-mm-thick layer of plastic was placed adjacent to the ^{235}U, between the uranium and a 15-mm-thick piece of molybdenum to be studied. Fast neutrons from the explosion produced fissions in the ^{235}U, resulting in a heating pulse for this material with a width of a fraction of a microsecond. Appropriate shielding between the source and the uranium absorbed slow neutrons that would have lengthened the heating pulse. The heating of the ^{235}U resulted in a pressure of about 45 million atmospheres, and the subsequent expansion of this material compressed the plastic to a thickness of 0.5 mm and drove a shock of 20 million atmospheres into the molybdenum.

Evacuated light pipes set at three different depths into the molybdenum gathered light as the shock broke through the free surfaces and transmitted it through massive shielding to phototubes 12 meters away. From these signals the shock velocity was determined.

The purpose of the plastic between the ^{235}U and the molybdenum was to slow down a very small fraction of the fast neutrons that traversed it and, after a fraction of a microsecond delay from the slowing-down process, to send a short burst of moderated neutrons through the molybdenum. The duration of the burst was less than one microsecond and consisted of a continuous velocity spectrum from about 40 mm/μs to several hundred mm/μs. The transmitted neutron velocity spectrum was measured by recording the arriving neutron intensity as a function of time using neutron detectors positioned 20 meters away. Useful resonances in the sample corresponded to neutron velocities from 250 to 500 mm/μs and the arrival times of these neutrons at the detectors spanned the interval from 40 to 80 microseconds after the short neutron pulse traversed the sample. Six pairs of dips in the velocity spectrum corresponded to large, well-known resonances in the stationary and shocked material. From each pair a value of the particle velocity was obtained. One such pair is shown in Fig. 1. The points are digitized readings of a neutron detector signal and show the absorption of neutrons by a well-known molybdenum resonance. The dip on the left corresponds to absorption by shocked material and that on the right to stationary material. The two-microsecond difference in arrival time of the neutrons corresponds to the derived particle velocity of 10.7 mm/μs. This velocity, combined with the shock velocity of 18.2 mm/μs as measured from light flashes, defines a pressure behind the shock of 20 Mbar and a corresponding density of 25 g/cm^3. The accuracy of the velocity measurements was about ± 5%, which is not as accurate as needed for a standard, but experience gained in this first measurement leads us to believe that an accuracy of ± 2% is possible.

III. CONCLUSION

We have demonstrated the feasibility of measuring simultaneously shock and particle velocities in molybdenum at a pressure of 20 Mbar. When these measurements are further refined, they will help define a standard relative to which other materials can be compared, in experiments similar to those developed in the U.S.S.R.

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FIGURE CAPTION

Fig. 1 Digitized data from a detector used to measure the neutron velocity spectrum in the region around one molybdenum resonance. This pair of dips represents the best data obtained in the particle velocity measurement. The solid lines represent least-squares fits of quadratics to the minima. The minimum on the right corresponds to the unshifted molybdenum resonance and the one on the left corresponds to the shifted resonance. The observed time difference of $\sim$ 2 μs for this pair of dips leads to a particle velocity of $\sim$ 10.7 mm/μs.

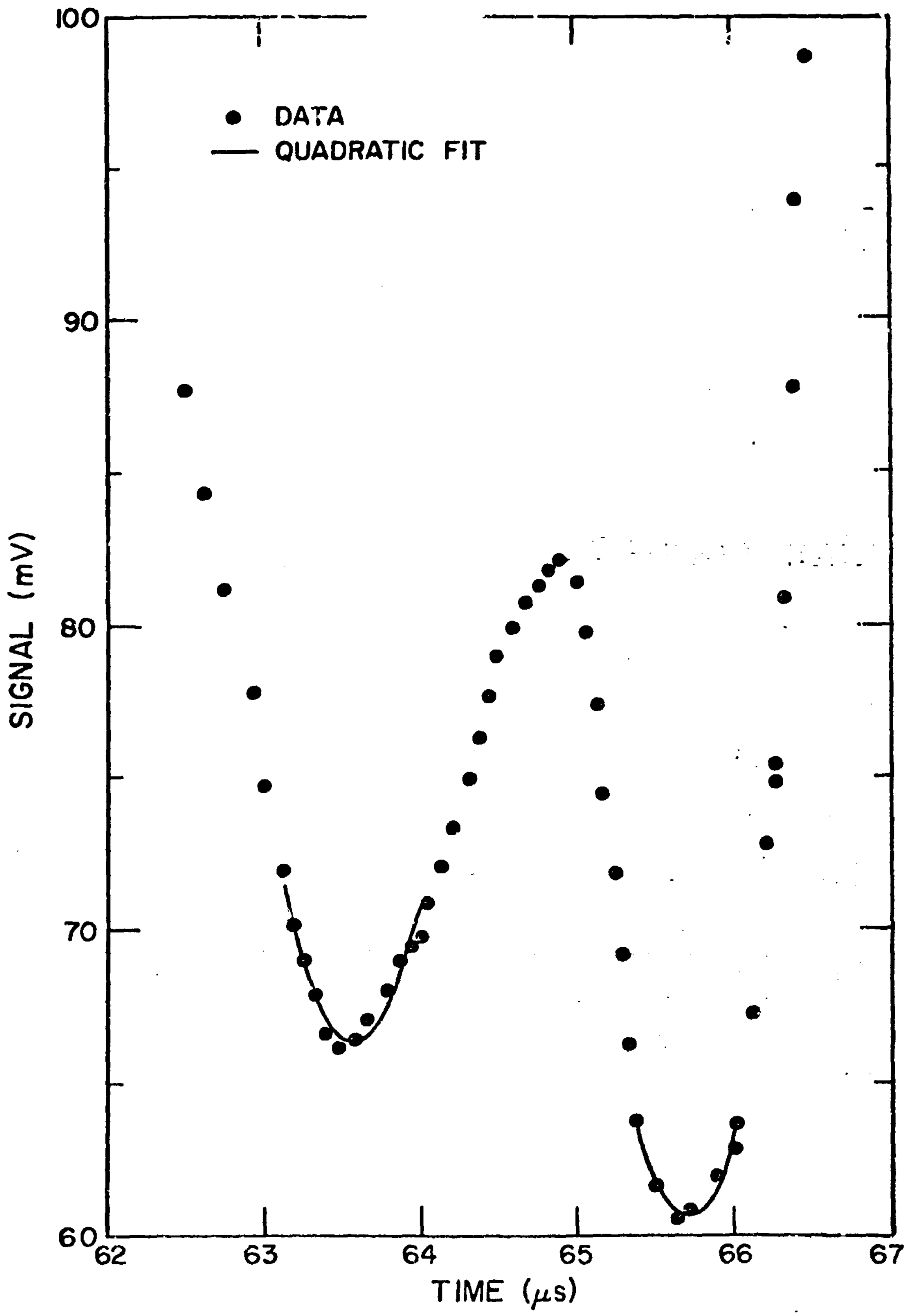
DATA
QUADRATIC FIT
SIGNAL (mV)
100
90
80
70
60
62
63
64
65
66
67
TIME (μs)

FIGURE 1