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SOUND VELOCITY OF CARBON AT HIGH PRESSURES⁺

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Résumé - Nous avons mesuré la vitesse du son dans le carbone comprimé par une onde de choc. Les résultats sont compatibles avec un modèle incluant l'existence de la phase diamant entre 0,8 et 1,4 Mbar.

<u>Abstract</u> - We have measured the sound velocity in shock compressed graphite. The data are consistent with a model of solid diamond from 0.8 to 1.4 Mbar.

INTRODUCTION

The carbon phase diagram is poorly known, even though this element is common, and its atomic structure is relatively simple. The high pressure and temperature of the graphite-liquid-vapor triple point have resulted in an uncertainty of the triple point temperature between 4200K /1/ and 4800K /2/. The diamond-liquid phase line has been presumed to have a negative pressure derivative on the basis of similarity to silicon and germanium /1/ and old shock wave data /3/, which was probably incorrect /4/. The similarity of liquid carbon to silicon and germanium may also be questioned, since the latter are normal liquid metals at atmospheric pressure, while fluid carbon has a resistivity above $10^{-3} \Omega$ -cm /5/.

Several theoretical phase diagrams for carbon have been presented. By use of electronegativity as a scaling parameter, Van Vechten has successfully constructed universal phase diagrams for group IV elements and binary alloys crystallizing in the diamond structure /6/. We show his results for carbon in Figure 1. Grover has done a more detailed thermodynamic analysis based on the Van Vechten phase diagram, and he concludes that diamond may not even be contiguous with the solid-liquid boundary /7/. Most recently, Yin and Cohen, using an ab initio pseudopotential theory, have cast doubt on the previous theoretical models /8/. The lack of p states in the carbon core and the relatively large beyond-valence orbitals combine to make diamond is even more compact than the nominally metallic close packed structures. The same arguments call into question the assumption that, since the diamond structure is open, the liquid must be denser.

Previous shock compression experiments on graphite in the pressure range 300-600 kbar show consistently that carbon collapses to a material with diamond density /4,9/. The presence of diamond crystallites in samples recovered from graphite shock compressed to 300 kbar for 1 µsec prove that the high density phase consists of at least small crystals of diamond /10/. However, although the Hugoniot for high density graphite crosses the melting line shown in figure 1 at accessable pressures, no indication of melting has been seen.

In a series of papers, the optical analyzer technique /ll/ has been used to measure the release wave velocity at high pressure for iron /l2/, tantalum /l3/, and aluminum /l4/. For these metals, the onset of melting is indicated by a dramatic decrease as pressure is increased in this velocity, from that characteristic of a longitudinal elastic wave to that of a bulk wave. Where this reduction in velocity

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has been compared with previously known phase boundaries, the agreement with equilibrium melting has been good /15/. We have chosen this technique to study the carbon phase diagram in the pressure range 800 kbar to 1.4 Mbar, and at temperatures of several thousand K.



Fig. 1 - Hypothetical phase diagram of carbon. The calculated Hugoniot, with our data points is indicated by the dashed line.

EXPERIMENTS

In these experiments we have chosen a pyrolytic graphite with an initial density of 2.20 gm/cc, and an orientation such that the shock propagation direction was perpendicular to the basal planes. The shocks were driven into the carbon targets by hitting them with thin iron or tantalum plates accelerated in a two-stage light gas gun. Iron was used for shock pressures below 1.2 Mbar, where the tantalum release velocity is not yet experimentally calibrated. A tantalum impactor was used for a high pressure experiment at 1.43 Mbar.

Bromoform was used as the optical analyzer material in contact with the graphite. The targets were cut so that several sample thickness could be used on each shot. The thermal radiation from the shock driven into the bromoform from each carbon thickness was recorded with photomultiplier and oscilloscope. By extrapolating the results to the carbon thickness required for the release to overtake the shock at the carbon-bromoform interface we calculated the velocity of the lead characteristic of the release wave at each shock pressure. These results are plotted in figure 2 with an estimated uncertainty of $\pm 3\%$ in sound velocity.



Fig. 2 - New sound velocity data for shock compressed graphite. For comparison we show calculated bulk and longitudinal velocities for diamond.

DISCUSSION

We show in figure 2 a calculated bulk sound velocity for solid diamond. This calculation is based on shock data measured by Pavlovskii /16/. With an initial density of 3.51 gm/cc, the zero pressure bulk velocity is 12 km/s, and the bulk modulus is 5.05 Mbar, close to McSkimin's ultrasonic results /17/. We have used the relationship $\rho\gamma$ =constant, commonly used for metals, with γ =1.1 at zero pressure We also plot several curves for the longitudinal wave velocity given /18/. constant Poisson's ratios. Although the zero pressure value of Poisson's ratio is about 0.1 /18/, the best fit to our high pressure data is 0.35, still a reasonable value for cubic solids. Our data are consistent with a longitudinal elastic wave in diamond, although the pressure dependence is slight. A steady increase in Poisson's ratio would explain the relatively low pressure dependence of the longitudinal velocity.

For comparison, we also show in figure 1 a calculated Hugoniot curve /4/ and the location of our data points. We see no evidence for a phase transition from the diamond structure. In order for us to miss the melting transition, the liquid carbon would need a bulk modulus twice that of diamond. Therefore, the melting line must have a near-zero or positive slope in the P-T plane.

The preliminary results presented here suggest that the electronegativity based phase diagram is incorrect for carbon. If true, the fluid in equilibrium with diamond may not be denser, and it may not be metallic. These conclusions are at least qualitatively in agreement with the pseudopotential based phase diagram. More sound velocity experiments are planned both at lower and higher pressures. Monte Carlo calculations, using the ab initio pseudopotential, could be particularly useful for determining the nature of high pressure fluid carbon.

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