LA-UR-02-2701
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Title: **MULTI-SCALE MODELLING OF BERYLLIUM: QUANTUM MECHANICS AND LASER-DRIVEN SHOCK EXPERIMENTS USING NOVEL DIAGNOSTICS**

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Submitted to:

MULTI-SCALE MODELLING OF BERYLLIUM: QUANTUM MECHANICS AND LASER-DRIVEN SHOCK EXPERIMENTS USING NOVEL DIAGNOSTICS*

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Ab initio quantum mechanics was used to construct a thermodynamically complete and rigorous equation of state for beryllium in the hexagonal and body-centred cubic structures, and to predict elastic constants as a function of compression. The equation of state agreed well with Hugoniot data and previously-published equations of state, but the temperatures were significantly different. The hexagonal / bcc phase boundary agreed reasonably well with published data, suggesting that the temperatures in our new equation of state were accurate. Shock waves were induced in single crystals and polycrystalline foils of beryllium, by direct illumination using the TRIDENT laser at Los Alamos. The velocity history at the surface of the sample was measured using a line-imaging VISAR, and transient X-ray diffraction (TXD) records were obtained with a plasma backlighter and X-ray streak cameras. The VISAR records exhibited elastic precursors, plastic waves, phase changes and spall. Dual TXD records were taken, in Bragg and Laue orientations. The Bragg lines moved in response to compression in the uniaxial direction. Because direct laser drive was used, the results had to be interpreted with the aid of radiation hydrodynamics simulations to predict the loading history for each laser pulse. In the experiments where there was evidence of polymorphism in the VISAR record, additional lines appeared in the Bragg and Laue records. The corresponding pressures were consistent with the phase boundary predicted by the quantum mechanical equation of state for beryllium. A model of the response of a single crystal of beryllium to shock loading is being developed using these new theoretical and experimental results. This model will be used in meso-scale studies of the response of the microstructure, allowing us to develop a more accurate representation of the behaviour of polycrystalline beryllium.

INTRODUCTION

Beryllium is one of the principal candidate materials for constructing the fuel capsule in ignition studies for inertial confinement fusion (ICF) (1). For this application, it is necessary to implode a hollow sphere of beryllium ~2 mm in diameter. In order to achieve ignition of the fuel, the implosion must be highly symmetric. However, beryllium exhibits strong elastic anisotropy, so a potential concern is the seeding of instabilities by spatial variations in the early acceleration history, resulting from inhomogeneities in the shock speed. One possible palliative is to drive the capsule with a first shock strong enough to cause melting, on the basis that that the liquid should not exhibit the anisotropy. However, a stronger shock in the capsule also causes greater heating of the fuel, making it harder to reach

a given compression and thus more difficult to ignite. It is therefore important for ICF to quantify anisotropy in shock propagation through polycrystalline beryllium, and to measure where shock melting occurs. Unfortunately, there are significant variations between different equations of state (EOS) and melt curves, and one would expect the constitutive behaviour of beryllium to vary considerably between microsecond time scales where models have been calibrated, and the nanosecond time scales relevant to ICF.

In this paper we describe some of our theoretical and experimental work on the shock response of beryllium on nanosecond time scales. We are pursuing a multi-scale approach, in that we have calculated and measured (some) properties of single crystals, and are using theoretical methods to predict the response of polycrystalline samples, with experiments for comparison.

THEORETICAL EQUATION OF STATE AND ELASTICITY

An ab initio quantum mechanical treatment was used to calculate the EOS and elasticity. The EOS was decomposed into a frozen-ion cold curve, thermal excitations of the electrons in the framework of the T=0 band structure, and phonon. The cold curve, band structure and dynamical matrix were calculated from the ground state wavefunctions of the outer electrons with respect to ions represented with ab initio pseudopotentials, as was done previously for silicon (2). This procedure can be applied independently to different crystal structures, resulting in an EOS for each. An equilibrium EOS can be deduced using Maxwell constructions. In the case of beryllium, the STP structure is hexagonal with a c/a ratio smaller than the ideal close-packed value, and on heating at 1 atm a phase transition occurs to body-centred cubic (bcc) just below melt (3). Since the hexagonal structure has an internal degree of freedom – the c/a ratio as well as the mass density – we performed a series of calculations to predict the variation of c/a with compression. These were consistent with the observed c/a ratio at STP (Fig. 1). Thermodynamically complete EOS were generated as rectangular tables of energy and pressure varying with mass density and temperature. Exchange and correlation in the electrons were modelled using the local density approximation (LDA) (4); as is usually found, the LDA overpredicts the binding energy of the crystal, hence underpredicts the p=0 lattice parameter by ~1%. As before, EOS were corrected to match the observed STP lattice parameter by adding a constant pressure (and corresponding energy varying linearly with volume) to the EOS. The same correction was made to predictions of the EOS in both hexagonal and bcc structures.

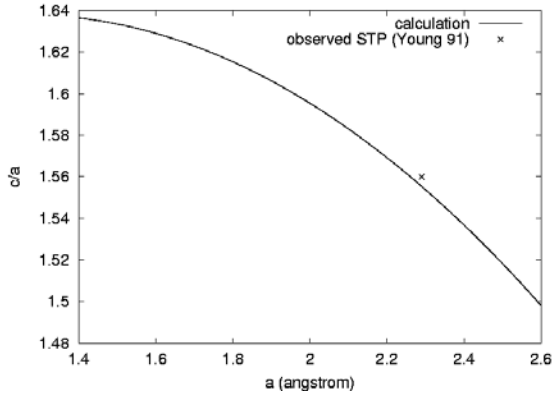

FIGURE 1. Variation of c/a ratio with compression for beryllium in the hexagonal structure.

Phase boundaries are usually very sensitive to the details of the theoretical procedure, and for beryllium the hexagonal/bcc boundary was not in particularly good agreement with the observed position. The bcc ground state energy was adjusted slightly to bring the 1 atm transition point into agreement; this led to a prediction of the pressure variation suggesting that the transition would occur for T=0 at ~45 GPa (Fig. 2). A transition in this pressure regime could affect the details of the shock response in the ICF capsule, and so is of potential interest.

The shock Hugoniot for beryllium was calculated by numerical solution of the Rankine-Hugoniot relations for the tabular EOS. In mechanical spaces, the predicted Hugoniot agreed well with gas gun data (5) and published empirical EOS (6) (Fig. 3). There was however a significant variation in the predicted temperature, which was not the case for the Hugoniot of aluminium (Fig. 2).

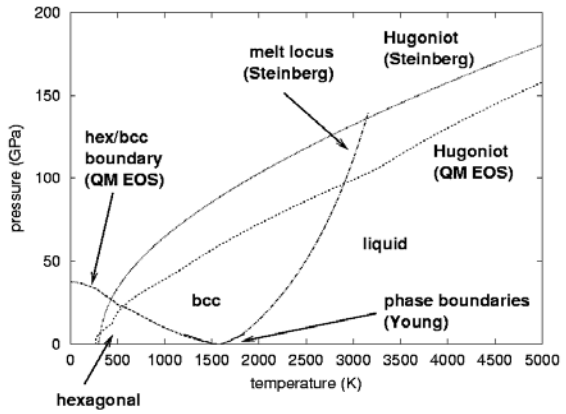

FIGURE 2. Phase diagram for beryllium.

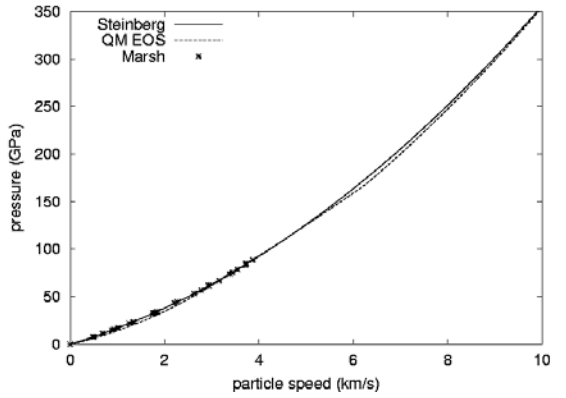

FIGURE 3. Shock Hugoniot from QM EOS, compared with gas gun data and empirical EOS.

The single-crystal elastic properties were predicted by performing electron ground state calculations with the lattice cell distorted from its equilibrium state. The variation in the stress components with strain were obtained directly from these calculations, and used to calibrate an analytic function.

LASER-DRIVEN SHOCK EXPERIMENTS

Experiments were performed using direct illumination by the TRIDENT laser (7) to drive a shock wave through a thin (~30μm) sample by surface ablation (8). The samples included single crystals cut parallel to the (0001) planes, and rolled foils. As reported previously (9), the experiments on single crystals included transient X-ray diffraction (TXD) as an *in situ* diagnostic of the response of the crystal lattice. Experiments on crystals and foils used an imaging VISAR-type velocimeter to measure the velocity history across a line on the opposite face to the drive beam (Fig. 4). Experiments were performed with shock pressures up to a few tens of gigapascals, using up to ~250 J of laser energy distributed evenly (compared with the sample thickness) over a focal spot 5 mm in diameter. One disadvantage of laser drive compared with flyer impact is that the relation between intensity history in the laser beam and pressure history in the ablated material is not straightforward, so the intensity history was recorded in each experiment, using a photodiode, and radiation hydrocode simulations were used to help interpret the data.

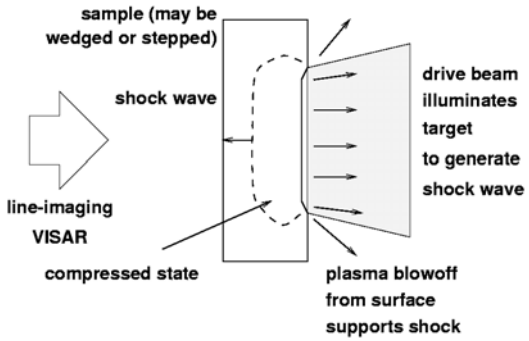

FIGURE 4. Schematic of shock generation by direct laser illumination.

The velocity records exhibited an elastic precursor, followed by the main plastic shock wave. The velocity history was constant over the image line, to the accuracy of the velocity reconstruction. For the (0001) crystals, the precursor had a rise time below the temporal resolution of the VISAR system (~2 ps). It was ~5 to 10 times stronger than one would expect from the published yield stress, which was inferred from experiments on ~microsecond time scales. In addition, it exhibited a slight deceleration after the initial maximum, before the main plastic wave reached the surface. (Fig. 5.)

Although the rocking curve of the crystals was ~2^{o} wide, compared with a field of view ~12^{o} for the TXD records, the dynamic range available on the recording media made it possible to interpret the records in terms of several distinct lines,

corresponding to discrete values of the lattice parameter. These values were superimposed on a background which presumably included contributions from regions in the sample where the lattice parameter varied continuously with position. With the help of comparisons with radiation hydrodynamic simulations of the experiments, identifications were proposed for each line. These included the elastic precursor, what was apparently a plastic decay of the precursor (i.e. the same compression, but in a state of isotropic rather than uniaxial stress), the main plastic wave, and additional lines at higher deviations in the Bragg angle which might be either transient regions of higher compression early in the laser pulse, or signals from lattice transformed to the bcc structure. It was possible to determine the position of the stronger lines to ~0.05°, and the weaker lines to ~0.1°. The uncertainty in the absolute lattice parameter was similar; the uncertainty in *differences* in lattice parameter was a few percent for the shock compressions achieved. (Fig.6.)

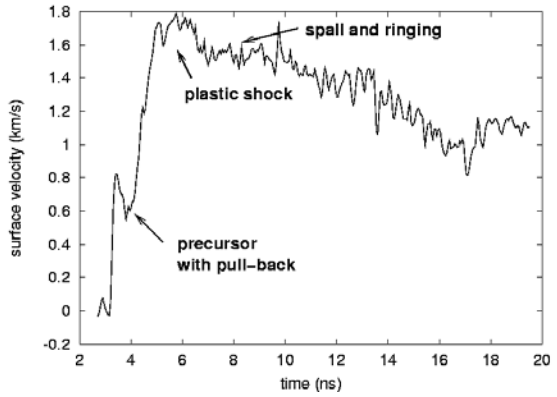

FIGURE 5. Example surface velocity record for an (0001) beryllium crystal, ~30 μm thick, driven to ~18 GPa (TRIDENT shot 12198).

In the experiments on beryllium foils, significant spatial variations were seen in the velocity history from the line VISAR, with spatial scales consistent with the grain size in the sample. The elastic precursor was slightly weaker than for the (0001) crystals, and had a rise time ~0.5 ns. (Fig. 7.)

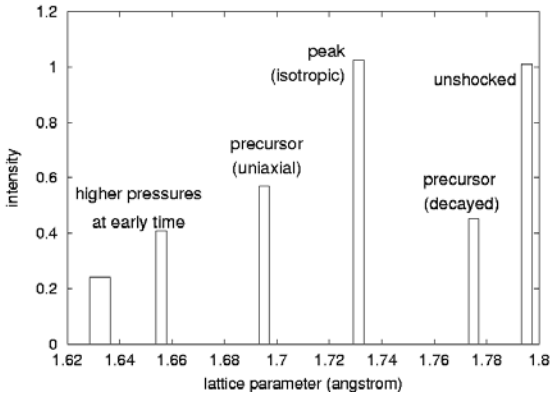

FIGURE 6. Example lattice parameters deduced from time-integrated TXD record for an (0001) beryllium crystal, ~30 μm thick, driven to ~30 GPa (TRIDENT shot 12202). The width of each bar represents its uncertainty; the nominal position is at the center of the bar. The height is the intensity of that signal: the integral of the amount of material of that lattice parameter over the duration of the X-ray pulse.

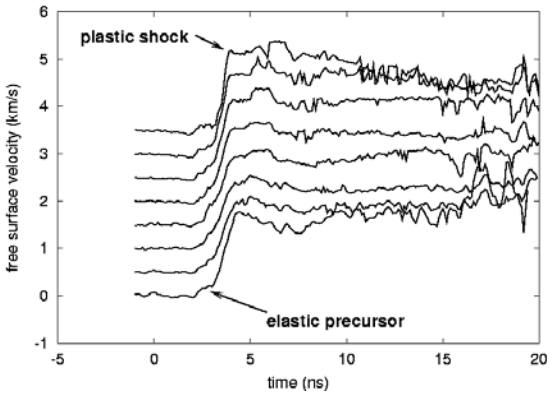

FIGURE 7. Example surface velocity records for a beryllium foil, ~30 μm thick, driven to ~13 GPa. The traces are from points 50 μm apart, with an offset applied to each for clarity. (TRIDENT shot 12182.)

Although the TRIDENT experiments were not designed for the measurement of EOS, Hugoniot states were deduced from both types of experiment. We experienced difficulty in deducing the shock speed with adequate accuracy, so states were inferred instead from the peak surface velocity and either the lattice parameter in the case of the single crystal experiments, or the time delay between the precursor and the plastic shock in the case of the foils. With the exception of the thinnest foils (~13 μm), where the velocity history was sensitive to fine details of the laser intensity history, the states

inferred were consistent with the published Hugoniot (Fig. 8). This suggests that there was no significant preheat caused by the use of the laser drive, and indicates that finite-strain corrections to the EOS (10) were not measurable in this regime.

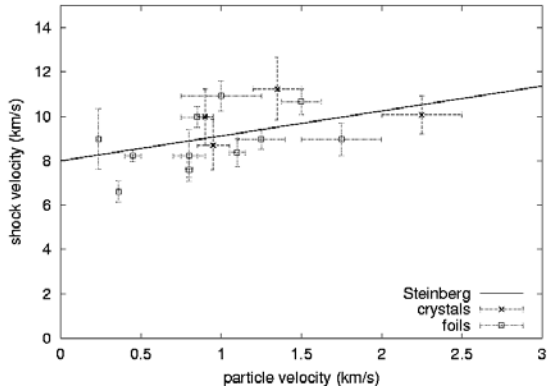

FIGURE 8. Shock Hugoniot states deduced from shock wave experiments on single crystals and polycrystal foils. Shock speed – particle speed is the space that exaggerates discrepancies in this data.

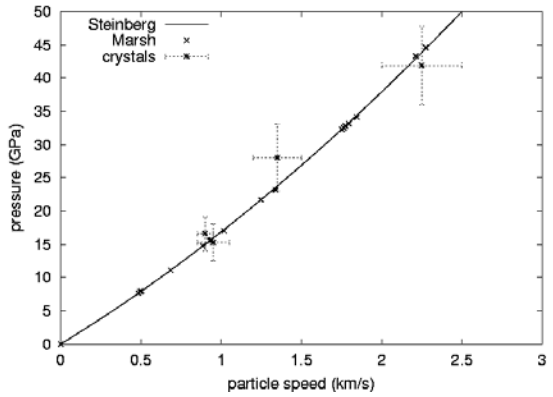

FIGURE 9. Shock Hugoniot states deduced from shock wave experiments on single crystals, compared with gas gun data.

More recently, we have performed flyer plate and quasi-isentropic compression experiments at the Z facility at Sandia National Laboratories. Pressures up to ~200 GPa were generated along the Hugoniot and isentrope. Detailed analysis and interpretation is still in progress, but it appears that the published EOS is still valid in these regions of state space, and there may be some evidence of melt on shocking or release.

DISCUSSION

We had hoped to be able to investigate melt in beryllium using TXD at TRIDENT. In the event, this did not prove possible because of a lack of beryllium crystals and the development work needed to perform TXD on experiments at over 100 GPa. However, we did demonstrate a significant difference in the elastic precursor between polycrystal foils and single crystals, which with further development work should allow us to place a constraint on the grain size to use for the ICF capsule so as to reduce the seeding of instabilities. The combination of TXD and line-imaging VISAR was valuable for investigating the detailed mechanical response and throwing light on underlying mechanisms.

The structure of the elastic precursor in (0001) crystals is interesting. The large magnitude compared with gas gun data is not surprising, since plastic flow is in principle time-dependent whether dominated by dislocations or twinning. The velocity pull-back may be caused by the crystal responding to the very large elastic strain by starting to deform plastically on the time scale of the experiment. This suggestion is supported by the observation from the TXD data that – as well as a line apparently corresponding to the uniaxial precursor – there was present a line corresponding to the same compression under an isotropic stress.

The TXD records also showed evidence of additional lines appearing for pressures over ~30 GPa. More work is needed to confirm the presence of these lines, but they could in principle be caused by material transforming to the bcc structure behind the shock wave. This would be in agreement with the two-phase QM EOS, although this EOS required some adjustment to reproduce the 1 atm hexagonal to bcc boundary, and therefore cannot be regarded as having the same degree of rigour as previous calculations of silicon and titanium.

The experiments described here were intended mainly to be scoping studies for the feasibility of TXD on beryllium crystals. In particular, their design and execution was not optimized for the determination of EOS points. Nevertheless, plausible Hugoniot states were deduced from the

data, with a scatter and uncertainty a few times that of gas gun data. We have since found it possible to reduce the uncertainty in our measurements of EOS points by roughly an order of magnitude, with some constraints on technique. It is certainly possible to make accurate measurements of quantities such as compressive and spall strengths, where the dominant uncertainty is usually the reproducibility of the microstructure, and relative measurements can be made more easily from the velocity records.

CONCLUSIONS

Quantum mechanical predictions were made of the equation of state (EOS) of beryllium, including a possible solid-solid phase transformation. The EOS was used to predict a shock Hugoniot; this was in reasonable agreement with gas gun data and an empirical EOS normalized to those data.

Experiments were performed using direct laser illumination to drive a shock into single crystal and polycrystal foil samples of beryllium. Pressures of up to a few tens of GPa were achieved with a couple of hundred Joules of light over a 5 mm spot. Line-imaging Doppler velocimetry and transient X-ray diffraction (TXD) were successfully fielded simultaneously on the single crystals, and surprisingly accurate lattice parameters were deduced from the TXD data, ~10 times narrower than the rocking curves.

Single crystals exhibited an elastic precursor several times stronger than would be predicted using gas gun strength data. There was also some structure to the precursor; possibly a plastic decay. In comparison, polycrystal foils exhibited a rather weaker precursor (though still stronger than would be expected from published strength models) with a significant rise-time.

Hugoniot states were deduced from both types of experiment, and were consistent with the gas gun data and empirical EOS.

ACKNOWLEDGMENTS

The quantum mechanical program was provided courtesy of the UK Carr-Parinello Consortium. Roger Kopp (Los Alamos National Laboratory) performed most of the radiation hydrodynamics simulations.

*This work was performed under the auspices of the U.S. Department of Energy under contract number W-7405-ENG-36.

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