Quasi-isentropic compression by ablative laser loading: Response of materials to dynamic loading on nanosecond time scales

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The TRIDENT laser was used to induce quasi-isentropic compression waves to ~15 GPa in samples of Si, by ablative loading using a laser pulse whose intensity increased smoothly over 2.5 ns. The intensity history of the pulse and the velocity history at the opposite surface of the sample were recorded. Experiments were performed using samples of two different thicknesses simultaneously, in which the evolution of the compression wave was clearly visible. Isentropic stress states deduced were consistent with the previously investigated response of Si to uniaxial loading. The ablative loading was simulated using radiation hydrodynamics, with different equations of state in the plasma and condensed regions and including elasticity in the solid Si. These calculations reproduced the measured velocity histories quite well, demonstrating that quasi-isentropic compression was induced with no preheat from the laser drive. Normal continuum behavior was demonstrated to hold below nanosecond time scales for isentropic compression waves, with no evidence for nonequilibrium effects in the crystal lattice. Details of the velocity history over about 10 GPa were reproduced less well, suggesting a deficiency in the model used for compressed Si, which may be consistent with recent theoretical predictions of uniaxial compression at high strain rates.

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I. INTRODUCTION

Dynamic loading along an isentropic path is a valuable complement to shock wave experiments for determining material properties such as the equation of state and dynamic plasticity, and for testing theoretical predictions of the behavior of condensed matter under compression. For a given pressure, the temperature on the isentrope is lower than in the shocked state, so isentropic compression explores different states than shocks. Approximately isentropic loading by uniaxial ramp-wave compression—generally referred to as isentropic compression experiments (ICEs)—has been demonstrated using electrical discharges [1,2] and expanding vapor clouds produced by the deposition of laser energy [3], on time scales of a few hundred nanoseconds. However, many of these experiments are used to calibrate models for explosively loaded systems in which the time increment used in continuum mechanics simulations may be 1 ns or less. Approximately isentropic loading is also necessary for efficient performance of inertially confined fusion implosions, by indirect (hohlraum) or direct (ablative) drive [4], with a duration of around 10 ns. Here we describe the use of ablative loading by temporally shaped laser pulses to induce quasi-isentropic loading, exploring the material response directly at time scales of order 1 ns. This technique is referred to here as laser-driven isentropic compression experiments (LICE). As with other ICE techniques, deviations from an isentropic path are to be expected as a result of uniaxial loading, because any plastic flow that occurs is thermodynamically irreversible—hence "quasi-isentropic."

II. EXPERIMENTAL METHOD

Samples of Si were subjected to ablative loading *in vacuo* by directing a pulse from one beam of the TRIDENT laser (a Nd:glass system) at one surface. The velocity history of the opposite surface was measured using line-imaging laser Doppler velocimetry (Fig. 1). The samples used were a few tens of micrometers thick by several millimeters across; samples were clamped in a reusable target holder. The Si was in the form of (100) crystals, 30 or 59 μm thick. The 30 μm samples had been coated with 0.1 μm of Al on the velocimetry side.

TRIDENT was operated in nanosecond mode with frequency doubling to 527 nm. The drive pulse comprised 13 elements of 180 ps (full width, half maximum), the intensity of each element being controlled independently. The relative intensities were chosen to produce an overall envelope which increased smoothly over ~2.5 ns. The total energy produced in each pulse was measured by diverting a few percent of the energy generated from an uncoated window to a calorimeter. The intensity history was measured similarly, using a photo-

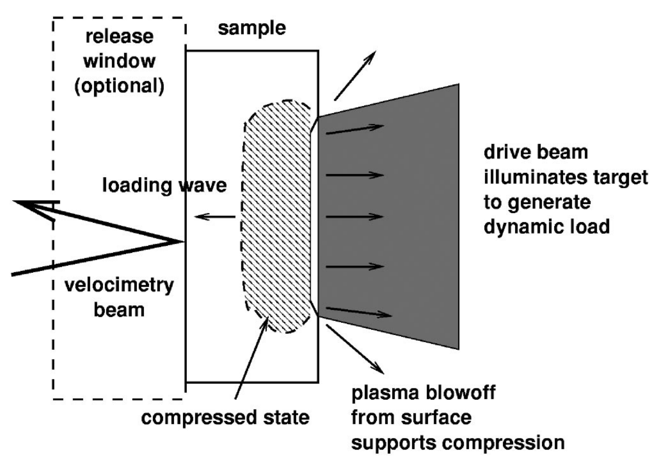

FIG. 1. Schematic of direct drive laser loading experiment (horizontal size exaggerated).

diode and a 6 GHz oscilloscope. Taking the response of the photodiode and cables into account, the temporal resolution was 70 ps. The measurement of total energy was calibrated by performing a series of shots with a second calorimeter inside the target chamber, in place of the target. As is generally found with high energy and high power lasers, the TRIDENT beams exhibit variations of the order of a factor of 2 in irradiance over spatial scales of tens of percent of the diameter of the beam. For experiments on dynamic material properties, it is highly desirable to have a uniform loading distribution in the transverse direction over a region much wider than the thickness of the sample. A diffractive optical element was used to distribute the beam more uniformly on large spatial scales at the expense of variations on spatial scales much smaller than the thickness of the sample. The irradiance distribution was determined by imaging the equivalent focal plane onto a charge-coupled device camera, and was verified by examining recovered samples, confirming that the optical element reduced the long wavelength variations to a few percent in irradiance and introduced variations of tens of percent on micrometer length scales. These peaks were not intense enough to cause localized preheating, and any resulting loading variations were compensated by plasma flow and wave interactions in the first micrometer or so of the sample, so the bulk of the sample experienced planar loading. The element used was a Fresnel zone plate, inserted between the final focusing lens ($f/8$) and the target, and designed to illuminate a spot 4 mm in diameter with 85% of the beam energy. If used at best focus, a small amount of undiffracted light formed a hotspot at the center of the spot. The beam was defocused by 18 mm to remove this hotspot at the cost of widening the rest of the spot and introducing a slight long-wavelength variation; fortuitously, the diffracted light exhibited a slight spatial variation at best focus (a central dip) and in fact the net effect of defocusing was to improve the uniformity of the irradiance over the central 2–3 mm (Figs. 2 and 3). No spatial variations in free surface velocity were observed from the drive. In this initial study, we concentrated on experiments sampling pressures up to $\sim$15 GPa, which were expected to remain isentropic over the sample thicknesses available. The laser energy per pulse was a few tens of joules.

The velocity measurement was by laser Doppler velocimetry of the Velocity Interferometry for Surfaces of Any Reflectivity (VISAR) type [5], using a probe laser operating at 660 nm and producing Gaussian pulses 100 ns long. The laser beam was spread onto a line on the sample, and the image of the line transferred to the VISAR interferometer, which was set to a velocity sensitivity of 425 m/s per fringe. The interference fringes were recorded using a streak camera (temporal resolution $\sim$200 ps in these experiments), along with timing pulses synchronized with the drive beam. The fringe motion was analyzed to infer the surface velocity history as a function of position across the sample; the uncertainty in the position of each fringe corresponded to a velocity uncertainty of around 30 m/s, reduced to around 10 m/s by averaging across fringes. Loading histories were identified as quasi-isentropic if the measured acceleration history was smooth. Additional experiments have been performed with a temporal resolution below 100 ps, demonstrating that smooth irradiance histories give smooth loading histories.

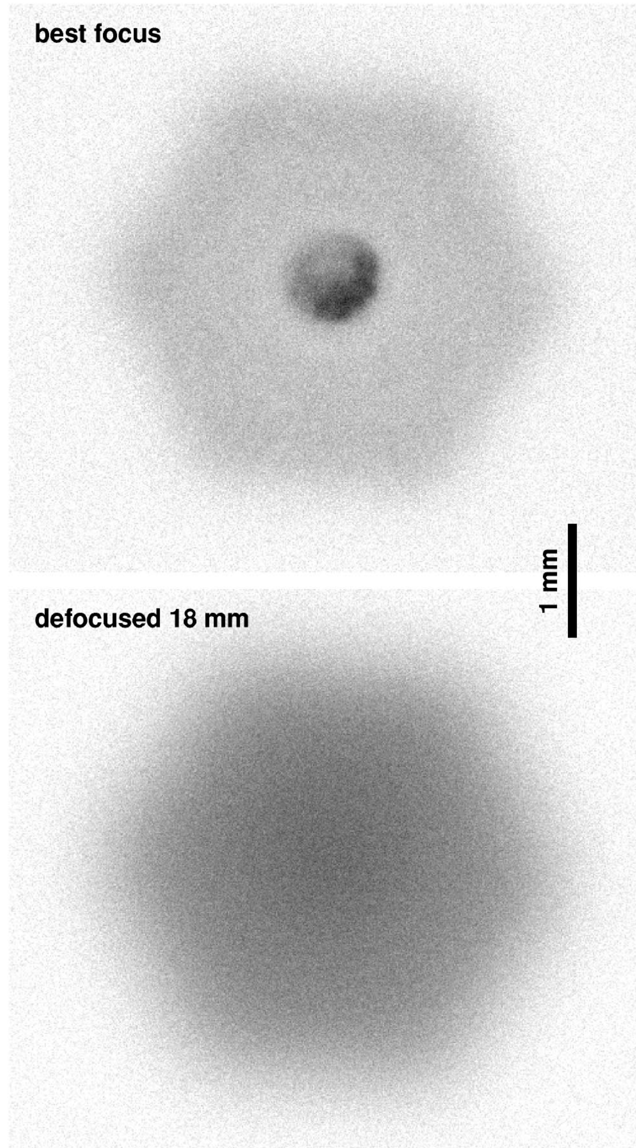

FIG. 2. Equivalent focal plane images of the spatial distribution of the laser drive spot, at best focus (top) and defocused to remove the central hotspot (bottom).

Twelve experiments were performed to investigate this technique: three on Al (two with a LiF release window), one on Be, five with a single sample of Si 59 μm thick, and three with pairs of Si samples. In some cases, the irradiance history was lost or the VISAR fringes were too weak for analysis. Simultaneously good irradiance and velocity records for Si were obtained on all the pairs and on three of the single samples. In all experiments in which a smoothly rising irradiance history was generated, the free surface velocity history exhibited smooth waves (Figs. 4–6).

The velocity records from simultaneously fired pairs of samples were analyzed to yield isentropes by calculating the difference in transit time as a function of particle velocity, as has been described previously [2]. The resulting stress-compression relations from different experiments were consistent with each other and with stress-strain relations pre-

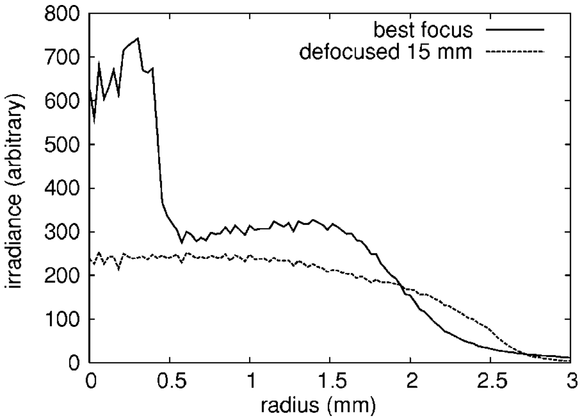

FIG. 3. Radial irradiance distribution (averaged azimuthally) for the laser drive spot, at best focus (solid) and defocused to remove the central hotspot (dashed).

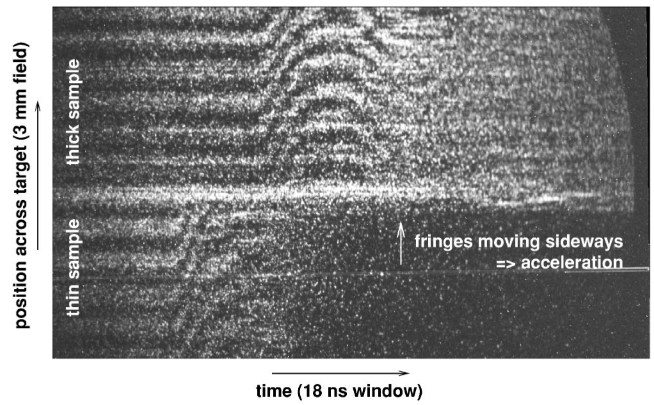

FIG. 5. Example line VISAR record for quasi-isentropic compression in Si (100) samples 30 and 59 μm thick (TRIDENT shot 15 020). Reflectivity was lost from the thin sample when the Al layer detached on deceleration. This record was from the highest drive energy attempted (62 J), which happened to give the weakest VISAR signal.

dicted assuming perfectly elastic response. At the peak stresses investigated in these experiments, the stress-strain behavior deviated little from linear elastic behavior (Fig. 7).

III. SIMULATIONS

In order to verify that the loading experienced by the sample was mechanical—with no significant heat conduction or preheat as a by-product of the ablative drive—simulations were performed using the measured irradiance history delivered by the laser. Radiation hydrodynamics was used to take account of the deposition of laser energy and its interaction with the resulting plasma plume, coupled with continuum mechanics to treat the response of the solid sample in reasonable detail. Separate simulations were performed using each technique, the pressure history predicted near the ablation surface by radiation hydrodynamics being used as an applied pressure boundary condition for continuum mechanics. The simulations were compared with the measured velocity histories.

In the regime explored by these experiments, laser ablation and dynamic loading can be simulated accurately by assuming three temperature hydrodynamics (ions, electrons, and radiation), including thermal conduction and radiation diffusion, and calculating the absorption of the laser energy in the expanding plasma cloud through the electrical conductivity [6]. This nonequilibrium model was needed only for material ablated by the laser ($\sim$1 μm of the solid); in this ablation regime, radiation hydrodynamics simulations and the microstructures of recovered samples indicate that nonequilibrium heating does not occur deeper than $\sim$1 μm from the drive surface. The remainder of the condensed phase was treated using nonradiative continuum mechanics, i.e., with an equation of state (EOS) and constitutive model only. Simulations were performed using the one-dimensional (1D) radiation hydrocode HYADES [7], which did not include material strength. A wide-ranging EOS for Si was taken from the SESAME database [8]; this was, however, inaccurate in compression, being based on the diamond structure in Si but with higher-pressure behavior estimated from the shock response of Ge. An EOS has been developed incorporating the diamond to body-centered tetragonal transition [9], which

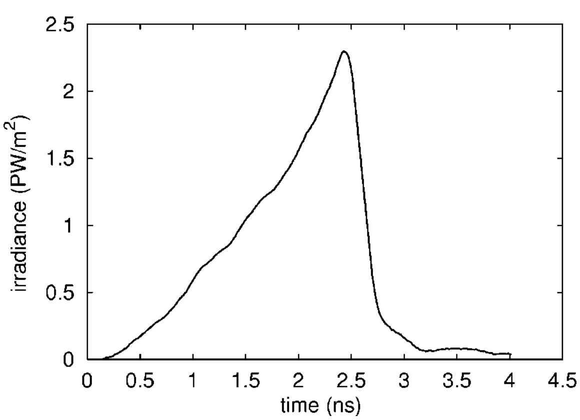

FIG. 4. Example of ramped laser irradiance history used to induce quasi-isentropic compression (TRIDENT shot 15 020).

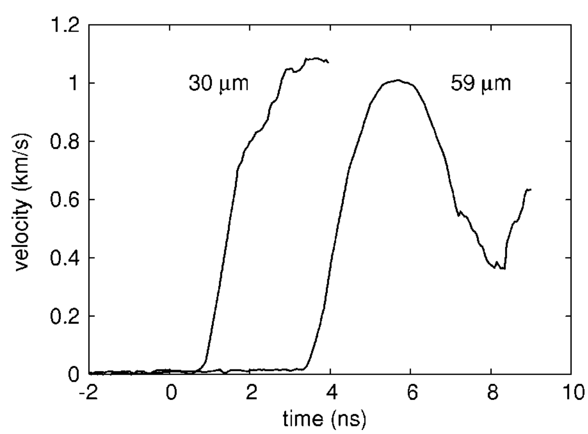

FIG. 6. Example surface velocity history for quasi-isentropic compression in Si (100) samples of two different thicknesses (TRIDENT shot 15 020). Lumpy structures near the peak velocity in the thin sample may be caused by noise in the VISAR record.

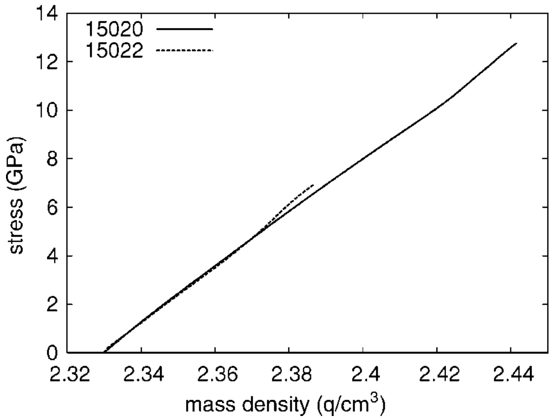

FIG. 7. Normal stress-compression relations obtained from experiments on pairs of Si samples of different thickness.

conversely does not cover high expansions or high temperatures but is more accurate in compression. Simulations were therefore set up in which a layer 1 μm thick at the surface illuminated by the laser was modeled using the SESAME EOS, and the rest of the sample was modeled with the more detailed solid EOS. Opacities for radiation diffusion were also taken from SESAME. Conductivities for laser deposition and heat conduction were calculated using the Thomas-Fermi ionization model [7,10]; this was found previously to be reasonably accurate for direct drive shock simulations on samples of a wide range of atomic numbers [6]. The pressure history 1 μm inside the drive surface was used as a time-dependent applied pressure boundary condition for the continuum mechanics simulations. The pressure history was predicted to increase fairly smoothly, though the simulations did exhibit some high frequency fluctuations caused by successive ablation of individual discrete cells of material (Fig. 8).

The spatial variations in irradiance introduced by the diffractive optical element were still within the range of validity previously found for the radiation hydrodynamics simulations. Thus nonhydrodynamic preheat effects would not be expected to occur in these experiments, in agreement with the velocimetry data.

Continuum mechanics simulations were performed using

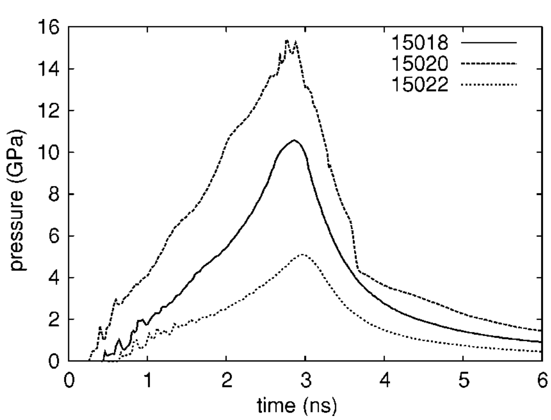

FIG. 8. Pressure-time histories predicted using radiation hydrodynamics, for shots on pairs of Si crystals. High frequency oscillations are numerical artifacts in the simulations.

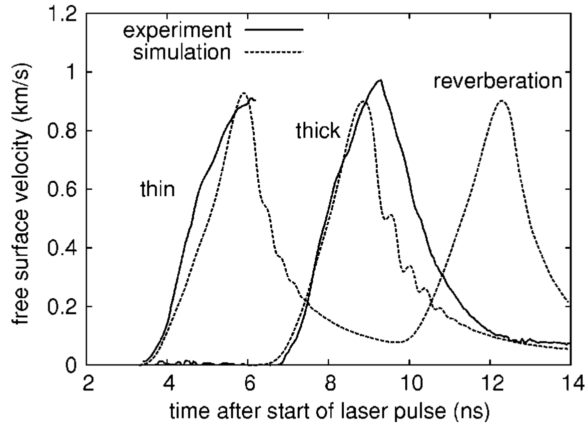

FIG. 9. Comparison between measured and calculated free surface velocity history for pair of Si (100) crystals irradiated simultaneously (TRIDENT shot 15 018). This agreement is within the uncertainty for measured velocity and drive energy.

the 1D hydrocode LAGC1D [11]. The response of Si to dynamic loading on nanosecond time scales is not well established [12], so for simplicity the constitutive behavior was treated using an elastic perfectly plastic model as it was easier to adjust the flow stress through a single parameter. The deviatoric shear modulus was 78.66 GPa.

For shots in which the sample was loaded to about 10 GPa or less, the free surface velocity history was reproduced quite well by the simulations (Fig. 9). For experiments exploring higher pressures, the observed velocity history exhibited a decrease in acceleration at around 0.7 ± 0.05 km/s, resulting in a smaller peak velocity than predicted in the simulations (Fig. 6). This deviation is likely to reflect a deficiency in the model for the response of solid Si, and may be connected with the previously observed anomalous behavior of Si under uniaxial loading [12,13]. In all cases, the loading history was clearly a smooth compression.

IV. DISCUSSION

The capability to perform experiments on different time and length scales allows the contributions to the dynamic response of competing phenomena to be separated, if they occur at different rates. Experiments exploring the regime covering a few nanoseconds–tens of micrometers–tens of gigapascals are valuable as different dislocation systems, twinning, and phase transitions in many materials may be active or frozen out [12]. The compression waves did not steepen into shocks over a sample thickness of several tens of micrometers. The energy in the drive pulse was well below the maximum available from TRIDENT (~250 J per beam), so it seems quite possible that this technique could be used to produce isentropic compression up to several tens of gigapascals in materials of higher atomic number and of a similar thickness, or to several hundred gigapascals using a smaller drive spot and thinner samples.

V. CONCLUSIONS

Temporally ramped laser pulses were found suitable for inducing quasi-isentropic compression waves by ablative loading of solid samples, allowing these LICE experiments to explore the material response on time scales of order nanoseconds, i.e., strain rates of order 10^8/s. For peak drive pressures of a few tens of gigapascals, samples can be several tens of micrometers thick before the compression wave steepens into a shock wave, allowing samples of similar size to grains in macroscopic engineering materials. Complete data on the evolution of a compression wave were obtained by irradiating samples of more than one thickness simultaneously. Coupled radiation hydrodynamics and continuum mechanics simulations of the material response were able to reproduce the free surface velocity history within the accuracy expected from the models, using the measured laser irradiance history. This accuracy is adequate for designing experiments and developing improved material models.

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