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Laser driven quasi-isentropic compression experiments (ICE) for dynamically loading materials at high strain rates

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We demonstrate the recently developed technique of laser driven isentropic compression (ICE) for dynamically compressing Al samples at high loading rates close to the room temperature isentrope and up to peak stresses above 100GPa. Upon analysis of the unloading profiles from a multi-stepped Al/LiF target a continuous path through Stress-Density space may be calculated. For materials with phase transformations ramp compression techniques reveals the location of equilibrium phase boundaries and provide information on the kinetics of the lattice re-ordering.

1. INTRODUCTION

The equation-of-state (EOS) of a material describes its response to applied stresses and temperatures and is widely used as an input into plasma and solid state physics codes. Methods for accurately measuring a materials path through stress-density space up to high levels of compression are therefore of considerable scientific interest. For materials with complicated phase maps compression can exhibit rate dependence if the timescale for a phase transformation is long relative to the compression rate. Therefore data needs to be amassed over a wide range of experimental conditions to build up a complete model of solid state transformations. To determine the kinetics of these transformations, for example, it is necessary to dynamically compress across phase boundaries and monitor the response of the material in a time resolved fashion.

Within this paper we describe a recently developed technique to shocklessly compress materials into the multi-Mbar pressure regime over tens of nanoseconds timescales. This isentropic compression experimental (ICE) platform allows for a single shot of continuous EOS points to be extracted close to the materials isentrope [1]. ICE-EOS measurements have several advantages over shock EOS experiments. Shock measurements give a single EOS point along the Hugoniot. Shock waves are associated with large jumps in temperature which greatly increases the thermal contribution to the pressure, and can cause melting of the material under study. In Al shock pressures above 1.2Mbar produce temperatures above ~4000K and melting of the sample. The isentrope generally lies to the compressive side of the Hugoniot in P-V space. In ICE experiments the shockless compression enables, for example, 1.2Mbar to be reached with temperatures of 500K [2]. The smaller amount of internal energy imparted into the material within the ICE platform allows for greater compression for comparable pressures on shock experiments. For materials which undergo polymorphic phase transformations, this ultra-fast ramp compression technique can be used to extract information about transformation kinetics and location of phase boundaries.

The shockless loading technique has been demonstrated with several drivers such as the magnetic pulse loading of the Sandia Z-machine [3], pillow impactors in gas guns facilities [4] and the chemical energy of high explosives [5]. The time scales for these experimental platforms range from 100's of ns to several microseconds. The experimental method for obtaining high quality EOS data close to the room temperature isentrope requires that a multi-step target must be compressed by a spatially planar pressure source. A direct Laser-driven shockless compression technique, developed for material strength measurements, has recently been demonstrated using the Omega laser up to peak stresses of 200 GPa [2, 6]. The planarity of the pressure drive reported in these experiments is limited by the performance of the current state-of-the-art laser focal spot smoothing techniques. The timescale for laser-driven ramp compression is up to two orders of magnitude faster than demonstrated on other

drivers. Therefore on comparing with data taken over vastly different timescales we can construct a picture of rate-dependent effects on, for example, material strength and phase transformation kinetics.

2. EXPERIMENTAL TECHNIQUE

2.1 Indirect Drive for EOS measurements

A method for extracting EOS information from shockless compression experiments as described by Aiden and Gupta [7] (illustrated in Fig. 1 [8]). The assumptions of the analysis are i) the sample always stays on the isentrope so that the EOS reduces to a single P_xr and ii) the ramp wave is a simple wave.

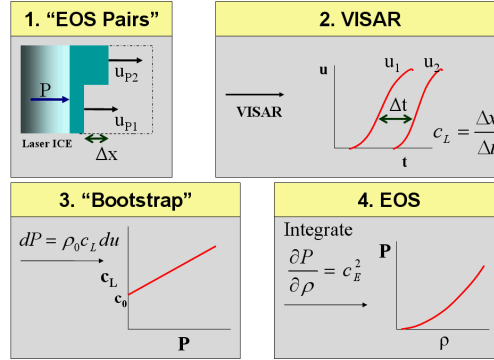


Fig 1. ICE EOS Lagrangian waveform analysis [7, 8]

An experiment which can employ the Lagrangian technique begins with uniform pressure wave applied to the step target with a step height Δx . We define the Lagrangian wave speed as $C_L(Up) = (p/\rho_0)C_E = \Delta x/\Delta t$, where Δt is the differential time difference between the two unloading step profiles for a given particle velocity, C_E is the Eulerian wave speed and ρ_0 is the material density. Figure 1 illustrates that given the step height we can determine C_L as a function of Up . Using the relationship, $dP = \rho_0 C_L dUp$, where P is the pressure we can bootstrap integrate along the $Up(t)$ curves to extract C_L as a function of pressure. Finally using the differential relationship $dP/d\rho = C_E^2$, we can integrate along the C_L vs P curve to obtain the isentropic relationship between pressure and density. It is also important that step heights be carefully designed so that wave reflections at the sample-window interface do not perturb the input wave history to the sample assembly and that wave perturbations due to the step do not perturb the measured profile for thicker steps. To extract $C_L(u)$ at either low pressures or when a window which is impedance matched to the sample can be used to allow direct observation of the insitu-velocity, this simple Lagrangian analysis can be used. However, at high pressures when strong release or interface effects are important, an iterative characteristic method outlined by Rothman [14] may be used.

In the experimental technique reported here laser energy is converted into a uniform x-ray drive which in turn is used to launch a pressure profile with a high degree of planarity over several hundred microns. This large planar region facilitates the use of multiple step targets all of which experience the same pressure source. The use of multiple thicknesses on a given target is necessary for improved accuracy and to determine deviations from simple wave behavior e.g. rate dependent strength effects. Using this technique we are able to extract single shot EOS data close to the room temperature isentrope of pure Al. The ICE-EOS package, as shown in Fig. 2 (a), consists of a Au hohlraum, a plastic reservoir followed by a vacuum gap and a double stepped Al target. Fifteen beams from the Omega laser at $0.35\mu\text{m}$ wavelength, containing a combined energy of 5kJ in a 2ns temporally flat pulse, are focused symmetrically onto the inner walls of the Au hohlraum (LEH: 1.7mm, diameter: 2.2mm, length: 1.7mm). This confined high-Z geometry results in a near blackbody distribution of thermal x-rays with uniform temperature gradients over a spatial region close to the diameter of the hohlraum. The hohlraum is attached to a $180\mu\text{m}$ thick 12% Br-doped polystyrene foil [$\text{C}_8\text{H}_6\text{Br}_2$]. The x-ray field within the hohlraum launches an ablatively driven shock through the foil. The initial region of planarity is expected to approach that of the diameter of the hohlraum and can extend over millimeters. The Bromine dopant absorbs high energy Au M-band x-rays generated within the hohlraum which otherwise could pre-heat the Al step sample. After breakout from the rear surface

shock heating and momentum cause the Br-CH to dissociate and unload across a 400 μm vacuum gap. Transit across the vacuum gap causes the mass density gradients along the target axis to relax as a function of distance from the original Br-CH / vacuum gap interface. The unloading Br-CH monotonically loads up against the Al sample and the imparted momentum launches a ramp stress wave through the material. The temporal profile of the compression wave may be shaped by varying the size of the vacuum gap, the density of the reservoir or the temperature within the hohlraum. Due to increase of sound speed with increasing pressure the ramp stress wave will eventually steepen up into a shock within the Al sample. This would result in a near instantaneous jump in entropy and off-isentropic compression (and possible target melting) would ensue. The maximum Al thickness is therefore designed to be less than the calculated shock-up thickness. In the experiments described here we used 10/20/30 μm Al steps into a well impedance matched LiF window. The thicknesses of the Al steps are chosen to compliment the target and irradiation conditions such that each step is shocklessly compressed.

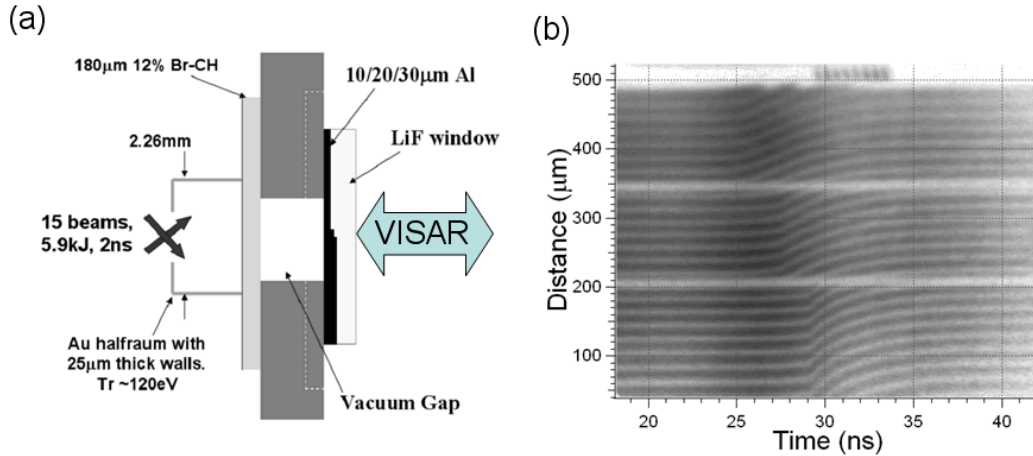


Fig. 2 (a) Indirect-drive Ice targets consist of a Low-Z reservoir, a vacuum gap and for EOS measurements a step target of the material of interest. (b) The output of the VISAR as recorded with a streak camera gives a temporal history of U_p and a spatial record of the applied pressure wave. Fringe movement is linearly proportional to the Al/LiF interface velocity. The time axis shown is relative to the incidence of laser radiation onto the inner walls of the Au hohlraum.

As the stress wave reaches the back surface of the Al target, the sample begins to accelerate into a well impedance matched LiF window. Probing through the LiF window, the time history of the Al/LiF interface acceleration is recorded with a line imaging velocity interferometer (VISAR) with two channels set at different sensitivities [9]. The time-resolved fringe movement recorded by a streak camera (see Fig. 2(a)) is linearly proportional to the velocity of the reflecting surface, which in this case is the Al-LiF interface. This allows for accurate measurement of the particle velocity (after taking into account the refractive index of the LiF window [10]) as a function of time. Shown in Fig. 1(b) is the streak camera output of the visar for the target conditions described in Fig. 1(a). The image provides spatial resolution at the target plane over $\sim 500\mu\text{m}$ and temporally resolution of the interferometer fringe displacement over a 30ns time window. We observe excellent planarity across the field of view with smooth ramp unloading from the 10, 20 and 30 μm Al samples at progressively later times. The velocity sensitivity (set by the resolving element within the visar) is $0.859067 \text{ km s}^{-1} \text{ fringe shift}^{-1}$. Using Fourier analysis [9] and after deconvolving the data for temporal and spatial distortions within the streak camera we can extract the time-resolved particle velocity (U_p) profile for each Al thickness. For all thicknesses we observe an elastic precursor with an associated particle velocity of $\sim 160 \text{ m/s}$ which is equivalent to a peak stress of $\sim 24 \text{ kbar}$ (using the Hugoniot relations for pure Al). This value is higher than the reported Hugoniot Elastic Limit (HEL) for pure Al ($\sim 4 \text{ kbar}$) and the enhancement is expected to reflect that the formation of dislocations required for the onset of plastic deformation is on a timescale long relative to the loading up time. The errors in the measurement which ultimately propagate through the Lagrangian analysis method described above are dominated by uncertainties in measuring a fringe shift. Using this technique we were able to measure a path through stress-density space with uncertainties are $\pm 5\%$ in stress and $\pm 2.5\%$ in density up to a peak stress of $\sim 100 \text{ GPa}$ [1]. These errors are dominated by uncertainties in measuring U_p ($\sim 0.1 \text{ km/s}$), time ($\sim 50 \text{ ps}$), step height ($\sim 1\%$), initial density (0.6%) but in total represents the most high quality, high pressure

isentropic data yet collected using laser drives and at the same time has unprecedented short loading time.

2.2 Direct Drive for Phase Transformation measurements

For EOS measurements a high degree of planarity is essential and therefore with the limitations of current beam smoothing techniques indirect drive and large laser facilities are required. However with moderate beam smoothing techniques on medium sized laser facilities we can explore the response of materials undergoing phase transformations when compressed over ultra-fast timescales. The location of phase boundaries are typically determined up to pressures of ~ 3 Mbar by various quasi-static experimental designs in which a sample is slowly heated and compressed while changes in the materials physical attributes such as electrical conductivity, sound speed, broadband reflectivity, lattice spacing, latent heat etc. are monitored to infer a phase transformation [11, 12]. For the majority of solid state phase transformations the relatively long timescale of these measurements ensure thermodynamic equilibrium between all phases. However, first order polymorphic phase transitions can be path and rate dependent and data needs to be amassed over a wide range of experimental conditions to build up a complete model of solid state transformations. To determine the kinetics of these transformations it is necessary to dynamically compress across phase boundaries and monitor the response of the material in a time resolved fashion.

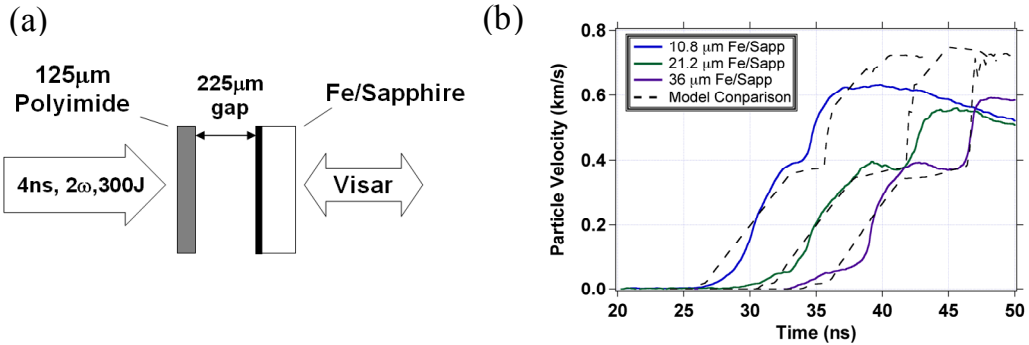


Fig 3. (a) Schematic of direct laser-driven ramp compression experiments on the Janus Laser facility. (b) Extracted Fe/Sapphire interface velocity profiles show signature of α - ϵ phase transformation at a higher velocity than predicted by multi-phase EOS hydrodynamic simulations.

In the laser driven shockless compression experiments reported here different thicknesses of Fe sample, directly coated onto a Sapphire window, are compressed to a peak longitudinal stress of ~ 21 GPa over 15ns. The Fe layer was coated onto the Sapphire window via electron beam deposition techniques. The target design, as shown in Fig. 3(a), consists of a 125 μ m polyimide [C₂₂H₁₀N₂O₅] foil followed by a 225 μ m vacuum gap and the Fe/Sapphire target. One beam of the Janus laser at the Lawrence Livermore National Laboratory delivered ~ 300 J in a 4ns square pulse at 527 nm onto the front surface of the polyimide foil which caused an ablatively driven shock to be launched into the material. A kinoform phase plate (KPP), inserted into the beamline to spatially smooth and shape the laser focal spot, generated a ~ 1 mm square planar ($\Delta I/I \sim 5\%$) region at the focal plane which contained an estimated 80% of the total drive energy, giving an on-target intensity of 3.8×10^{12} W/cm². After breakout from the rear surface shock heating and momentum cause the polyimide to unload across a vacuum gap. Transit across the vacuum gap causes the mass density gradients along the target axis to relax as a function of distance from the original reservoir - vacuum gap interface. The unloading reservoir material monotonically stagnates on the Fe sample, launching a ramp compression wave into the material.

Following the room temperature isentrope the α - ϵ solid-solid phase transformation in Fe is expected to occur at ~ 12 GPa [11]. As the ramp compression applied to the Fe reaches the pressure at which the phase transformation takes place the material undergoes a change in the sound speed [16]. As a result a multi-wave velocity feature is recorded on the visar (starting at an Fe/Sapphire interface velocity of ~ 0.4 km/s). The loading history at the front surface of the Fe sample can be determined by replacing the Fe/Sapphire target with an Al/LiF target. The particle velocity history of the Al into the well impedance matched LiF allows for the compression source to be determined via a back-integration

technique using the time dependent particle velocity at the rear surface as an input [13]. Using this pressure input we model the wave propagation in the Fe/Sapphire sample via a 1-D hydrodynamics code coupled to a multi-phase EOS model. The predicted outputs for three different thicknesses are shown in Fig. 3(b). We observe higher velocities for the onset of the multi-wave velocity features (associated with the phase transformation) experimentally which suggests there are kinetic effects at these high loading rates which cause deviations from the equilibrium phase EOS.

4. CONCLUSIONS

We have demonstrated a technique for shocklessly compressing materials with a high degree of planarity to over 100GPa. Upon analysis of the unloading profiles from a multi-stepped target a continuous path through Stress-Density space may be calculated [1].

For materials with many phases along the isentrope, wave profile allows us to locate the phase boundary, provides information on the kinetics of the phase transformation, and provides information on the relative change in sound speed between phases [16].

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